



(19)

(11) Publication number: **2000**

Generated Document.

**PATENT ABSTRACTS OF JAPAN**(21) Application number: **11200694**(51) Intl. Cl.: **H01B 1/06 C08F 2/44 C08F2  
3/24 C08L 83/04 H01G 9/02  
H01M 6/18 H01M 10/40**(22) Application date: **14.07.99**

(30) Priority: <b>05.08.9826.11.98 JPJP 1022214810336168</b>	(71) Applicant: <b>SONY CORP</b>
(43) Date of application publication: <b>11.08.00</b>	(72) Inventor: <b>NODA KAZUHIRO HORIE TAKESHI YASUDA TOSHIKAZU</b>
(84) Designated contracting states:	(74) Representative:

**(54) ELECTROLYTE  
COMPOSITION,  
ELECTROLYTE, ITS  
MANUFACTURE, AND  
BATTERY USING IT**

(57) Abstract:

**PROBLEM TO BE SOLVED:** To provide an electrolyte composition with high ionic conductivity, an electrolyte, its manufacturing method, and a battery using it.

**SOLUTION:** A negative electrode 11 and a positive electrode 12 are faced through an electrolyte 13. The electrolyte 13 is prepared by polymerizing an electrolyte composition containing a compound having ether linkage and a cross-linkable functional group, a siloxane derivative, and a lithium salt. The compound having ether linkage and the cross-linkable functional group forms three-dimensional net

structure, and the siloxane and the lithium salt exist in the net structure. High film forming capability and strong film strength are maintained and at the same time ionic conductivity is heightened.

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## PATENT ABSTRACTS OF JAPAN

(11)Publication number : 2000-222939

(43)Date of publication of application : 11.08.2000

(51)Int.Cl.

H01B 1/06  
C08F 2/44  
C08F299/00  
C08K 3/24  
C08L 83/04  
H01G 9/025  
H01G 9/028  
H01M 6/18  
H01M 10/40

(21)Application number : 11-200694

(71)Applicant : SONY CORP

(22)Date of filing : 14.07.1999

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(30)Priority

Priority number : 10222148  
10336168

Priority date : 05.08.1998  
26.11.1998

Priority country : JP

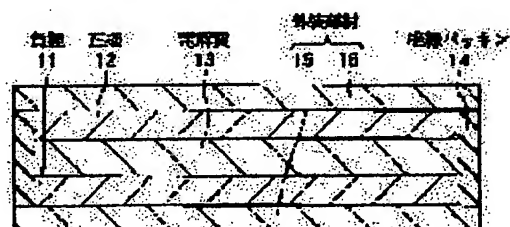
JP

(54) ELECTROLYTE COMPOSITION, ELECTROLYTE, ITS MANUFACTURE, AND BATTERY USING IT

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## LEGAL STATUS

[Date of request for examination]

[Date of sending the examiner's decision of rejection]

[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]

[Date of final disposal for application]

[Patent number]

[Date of registration]

[Number of appeal against examiner's decision of rejection]

[Date of requesting appeal against examiner's decision of rejection]

[Date of extinction of right]

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**Title:** JP2000222939A2: ELECTROLYTE COMPOSITION, ELECTROLYTE, MANUFACTURE, AND BATTERY USING IT

**Country:** JP Japan

**Kind:** A2 Document Laid open to Public inspection I

**Inventor:** NODA KAZUHIRO;  
HORIE TAKESHI;  
YASUDA TOSHIKAZU;

**Assignee:** SONY CORP  
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**Published / Filed:** 2000-08-11 / 1999-07-14

**Application Number:** JP1999000200694

**IPC Code:** [H01B 1/06](#); [C08F 2/44](#); [C08F 299/00](#); [C08K 3/24](#); [C08L 83/04](#);  
[H01G 9/025](#); [H01G 9/028](#); [H01M 6/18](#); [H01M 10/40](#);

**Priority Number:** 1998-11-26 JP1998000336168

**Abstract:**

PROBLEM TO BE SOLVED: To provide an electrolyte composition with high ionic conductivity, an electrolyte, its manufacturing method, and a battery using it.

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**INPADOC Legal Status:** None [Get Now: Family Legal Status Report](#)

**Designated Country:** CN EP KR US DE FR GB

**Family:** [Show 6 known family members](#)

**Other Abstract Info:** CHEMABS 132(11)145516B CHEMABS 133(13)180334H CHEMABS 133(13)180334H DERABS C2000-237367





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(19) 日本国特許庁 (J P)

(12) 公開特許公報 (A)

(11) 特許出願公開番号

特開2000-222939

(P2000-222939A)

(43) 公開日 平成12年8月11日 (2000.8.11)

(51) Int. CL <sup>7</sup>	識別記号	F I	特開2000-222939A (参考)
H 0 1 B 1/06		H 0 1 B 1/06	A
C 0 8 F 2/44		C 0 8 F 2/44	C
299/00		299/00	
C 0 8 K 3/24		C 0 8 K 3/24	
C 0 8 L 83/04		C 0 8 L 83/04	

審査請求 未請求 請求項の数52 O L (全 18 頁) 最終頁に続く

(21) 出願番号 特願平11-200694

(22) 出願日 平成11年7月14日 (1999.7.14)

(31) 優先権主張番号 特願平10-222148

(32) 優先日 平成10年8月5日 (1998.8.5)

(33) 優先権主張国 日本 (J P)

(31) 優先権主張番号 特願平10-336168

(32) 優先日 平成10年11月26日 (1998.11.26)

(33) 優先権主張国 日本 (J P)

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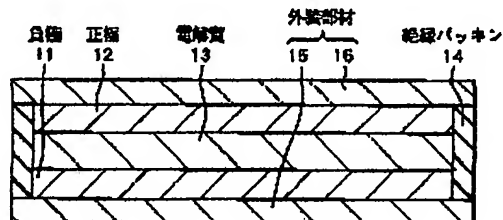
(74) 代理人 100083785  
弁理士 藤島 洋一郎

(54) 【発明の名称】 電解質用組成物ならびに電解質およびその製造方法およびそれを用いた電池

(57) 【要約】

【課題】 高いイオン導電率を有する電解質用組成物ならびに電解質およびその製造方法およびそれを用いた電池を提供する。

【解決手段】 電解質13を介して負極11と正極12とが対向して配置されている。電解質13は、エーテル結合および架橋可能な官能基を有する化合物とシロキサン誘導体とリチウム塩とを含む電解質用組成物を重合させたものにより構成されている。エーテル結合および架橋可能な官能基を有する化合物は重合により三次元網目構造を形成し、その間にシロキサン誘導体とリチウム塩が存在している。よって、高い成膜性および強い膜強度を保持しつつ、イオン伝導率を高くすることができる。



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## 【特許請求の範囲】

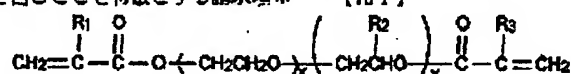
【請求項1】 エーテル結合および架橋可能な官能基を有する架橋可能な化合物と、

高分子化合物と、

電解質塩を含むことを特徴とする電解質用組成物。

【請求項2】 前記架橋可能な化合物は、架橋可能な官能基を少なくとも1つ有する1種または2種以上の化合物を含むことを特徴とする請求項1記載の電解質用組成物。

【請求項3】 前記架橋可能な化合物は、架橋可能な官能基が配されずかつエーテル結合を含む自由末端を少なくとも1つ有する化合物を含むことを特徴とする請求項\*



(式中、 $\text{R}_1$ 、 $\text{R}_2$  および  $\text{R}_3$  は水素原子またはアルキル基を表し、 $x$  および  $y$  は  $x \geq 1$  かつ  $y \geq 0$  または  $x \geq 0$  かつ  $y \geq 1$  の整数である。)

【請求項7】 前記架橋可能な化合物は、更に、下記の化2で表されるモノエステル化合物を含むことを特徴とする請求項6記載の電解質用組成物。

## 【化2】

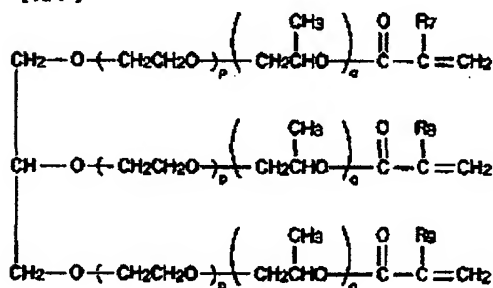


(式中、 $\text{R}_4$ 、 $\text{R}_5$  および  $\text{R}_6$  は水素原子またはアルキル基を表し、 $x$  および  $y$  は  $x \geq 1$  かつ  $y \geq 0$  または  $x \geq 0$  かつ  $y \geq 1$  の整数である。)

【請求項8】 前記ジエステル化合物に対する前記モノエステル化合物の重量比(モノエステル化合物/ジエステル化合物)は、0よりも大きく5、0以下の範囲内であることを特徴とする請求項7記載の電解質用組成物。

【請求項9】 前記架橋可能な化合物は、下記の化3で表されるトリエステル化合物を含むことを特徴とする請求項5記載の電解質用組成物。

## 【化3】



(式中、 $\text{R}_7$ 、 $\text{R}_8$  および  $\text{R}_9$  は水素原子またはアルキル基を表し、 $p$  および  $q$  は  $p \geq 1$  かつ  $q \geq 0$  または  $p \geq 0$  かつ  $q \geq 1$  の整数である。)

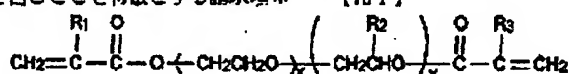
\* 1 記載の電解質用組成物。

【請求項4】 前記架橋可能な化合物は、エステル化合物を含むことを特徴とする請求項1記載の電解質用組成物。

【請求項5】 前記架橋可能な化合物は、モノエステル化合物、ジエステル化合物およびトリエステル化合物のうちの少なくとも1種を含むことを特徴とする請求項4記載の電解質用組成物。

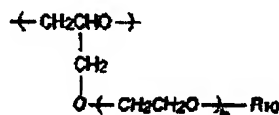
【請求項6】 前記架橋可能な化合物は、下記の化1で表されるジエステル化合物を含むことを特徴とする請求項5記載の電解質用組成物。

## 【化1】



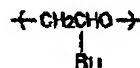
【請求項10】 前記架橋可能な化合物は、下記の化4で表される構成単位と、下記の化5で表される構成単位とを含む共重合体であることを特徴とする請求項1記載の電解質用組成物。

## 【化4】



(式中、 $\text{R}_{10}$  はアルキル基、アルケニル基、シクロアルキル基、アリール基、アラルキル基およびテトラヒドロピラニル基のうちの少なくとも1種を表し、 $h$  は  $1 \leq h \leq 12$  の整数である。)

## 【化5】



(式中、 $\text{R}_{11}$  は活性水素基および不飽和二重結合を有する官能基のうちの少なくとも一方、または活性水素基および不飽和二重結合を有する官能基のうちの少なくとも一方並びに水素原子、アルキル基、アルケニル基、シクロアルキル基およびアリール基のうちの少なくとも1種を表す。)

【請求項11】 前記架橋可能な化合物に対する前記電解質塩の割合は、架橋可能な化合物におけるエーテル結合単位のモル数に対する電解質塩のモル数の割合(電解質塩のモル数/架橋可能な化合物におけるエーテル結合単位のモル数)で、0.0001以上5以下の範囲内であることを特徴とする請求項1記載の電解質用組成物。

【請求項12】 前記高分子化合物は、下記の化6で表されるシロキサン誘導体を含むことを特徴とする請求項1記載の電解質用組成物。

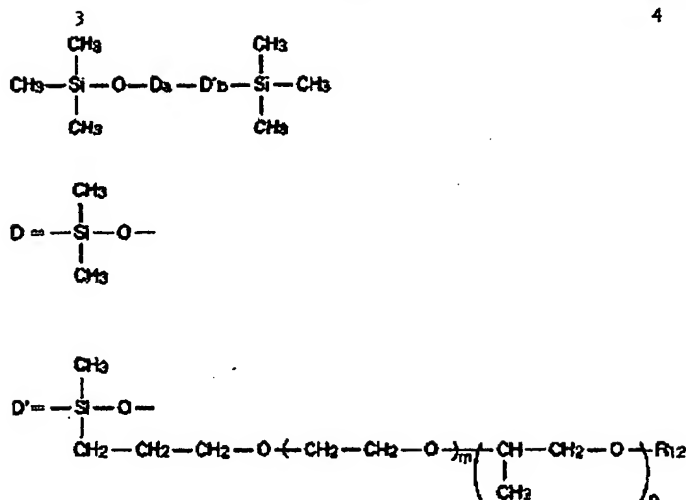
## 【化6】



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(式中、aは0から100の整数を表し、bは1から100の整数を表し、mは0から100の整数を表し、nは0から100の整数を表し、R<sub>12</sub>は水素原子またはアルキル基を表す。なお、b>1のときb個のD'は同じであっても異なってもよい。また、D'およびR<sub>12</sub>に含まれる水素原子はハロゲン原子で置き換えられていてもよい。)

【請求項13】 前記シリキサン誘導体に対する前記電解質塩の割合は、シリキサン誘導体におけるエーテル結合単位の数に対する電解質塩の数の割合（電解質塩のモル数/シリキサン誘導体におけるエーテル結合単位の数）で、0.0001以上5以下の範囲内であることを特徴とする請求項12記載の電解質用組成物。

【請求項14】 前記シリキサン誘導体の100重量部に対して前記架橋可能な化合物を10重量部以上1000重量部以下の範囲内で含むことを特徴とする請求項12記載の電解質用組成物。

【請求項15】 前記シリキサン誘導体の平均分子量は100000以下であることを特徴とする請求項12記載の電解質用組成物。

【請求項16】 前記電解質塩は、リチウム塩であることを特徴とする請求項1記載の電解質用組成物。

\* 【請求項17】 エーテル結合および架橋基を有する化合物が架橋基において架橋された化合物と、高分子化合物と、

20 電解質塩とを含むことを特徴とする電解質。

【請求項18】 前記架橋された化合物は、少なくとも1つの架橋基を有する1種または2種以上の化合物が架橋基において架橋された構造を有することを特徴とする請求項17記載の電解質。

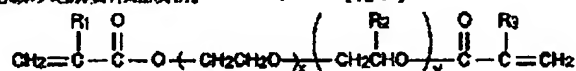
【請求項19】 前記架橋された化合物は、エーテル結合を含む自由末端鎖を有することを特徴とする請求項17記載の電解質。

【請求項20】 前記架橋された化合物は、エステル化合物が架橋基において架橋された構造を有することを特徴とする請求項17記載の電解質。

30 【請求項21】 前記架橋された化合物は、モノエステル化合物、ジエステル化合物およびトリエステル化合物のうちの少なくとも1種が架橋基において架橋された構造を有することを特徴とする請求項20記載の電解質。

【請求項22】 前記架橋された化合物は、下記の化7で表されるジエステル化合物が架橋基において架橋された構造を有することを特徴とする請求項21記載の電解質。

【化7】



(式中、R<sub>1</sub>、R<sub>2</sub>およびR<sub>3</sub>は水素原子またはアルキル基を表し、xおよびyはx≥1かつy≥0またはx≥0かつy≥1の整数である。)

【請求項23】 前記架橋された化合物は、更に、下記の化8で表されるモノエステル化合物が架橋基において架橋された構造を有することを特徴とする請求項22記載の電解質。

【化8】



(式中、R<sub>4</sub>、R<sub>5</sub>およびR<sub>6</sub>は水素原子またはアルキル基を表し、XおよびYはX≥1かつY≥0またはX≥0かつY≥1の整数である。)

50 【請求項24】 前記架橋された化合物は、ジエステル化合物が架橋基において架橋された構造とモノエステル

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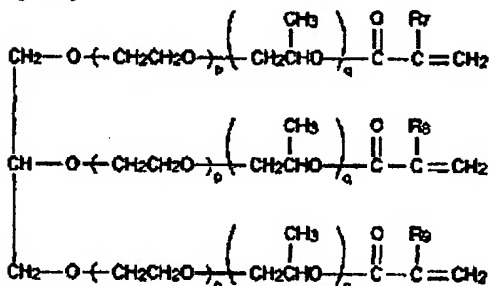
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化合物が架橋基において架橋された構造とを、ジエステル化合物に対するモノエステル化合物の重量比（モノエステル化合物／ジエステル化合物）において0よりも大きく5.0以下の範囲内で有することを特徴とする請求項23記載の電解質。

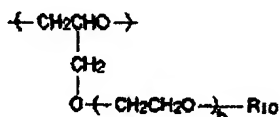
【請求項25】 前記架橋された化合物は、下記の化9で表されるトリエステル化合物が架橋基において架橋された構造を有することを特徴とする請求項21記載の電解質。

【化9】



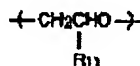
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\*20



（式中、 $\text{R}_{10}$ はアルキル基、アルケニル基、シクロアルキル基、アリール基、アラルキル基およびテトラヒドロピラニル基のうちの少なくとも1種を表し、 $h$ は $1 \leq h \leq 12$ の整数である。）

【化11】



（式中、 $\text{R}_{11}$ は活性水素基および不飽和二重結合を有する官能基のうちの少なくとも一方、または活性水素基および不飽和二重結合を有する官能基のうちの少なくとも一方並びに水素原子、アルキル基、アルケニル基、シク

\*（式中、 $\text{R}_1$ 、 $\text{R}_2$ および $\text{R}_3$ は水素原子またはアルキル基を表し、 $p$ および $q$ は $p \geq 1$ かつ $q \geq 0$ または $p \geq 0$ かつ $q \geq 1$ の整数である。）

【請求項26】 前記架橋された化合物は、下記の化10で表される構成単位と下記の化11で表される構成単位とを含む共重合体が架橋基において架橋された構造を有することを特徴とする請求項17記載の電解質。

【化10】

ロアルキル基およびアリール基のうちの少なくとも1種を表す。）

【請求項27】 前記架橋された化合物に対する前記電解質塩の割合は、架橋された化合物におけるエーテル結合単位のモル数に対する電解質塩のモル数の割合（電解質塩のモル数／架橋された化合物におけるエーテル結合単位のモル数）で、0.0001以上5以下の範囲内であることを特徴とする請求項17記載の電解質。

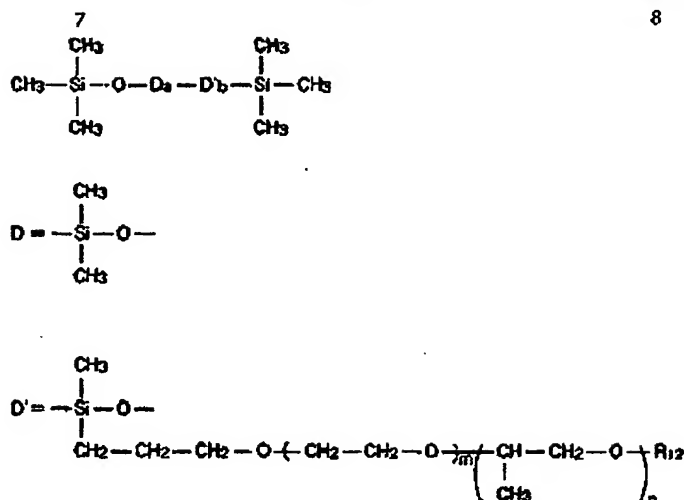
【請求項28】 前記高分子化合物は、下記の化12で表されるシロキサン誘導体を含むことを特徴とする請求項17記載の電解質。

【化12】

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(式中、aは0から100の整数を表し、bは1から100の整数を表し、mは0から100の整数を表し、nは0から100の整数を表し、R<sub>12</sub>は水素原子またはアルキル基を表す。なお、b>1のときb個のD'は同じであっても異なってもよい。また、D'およびR<sub>12</sub>に含まれる水素原子はハロゲン原子で置き換えられていてもよい。)

【請求項29】 前記シロキサン誘導体に対する前記電解質塩の割合は、シロキサン誘導体におけるエーテル結合単位のモル数に対する電解質塩のモル数の割合（電解質塩のモル数/シロキサン誘導体におけるエーテル結合単位のモル数）で、0.0001以上5以下の範囲内であることを特徴とする請求項28記載の電解質。

【請求項30】 前記シロキサン誘導体の100重量部に対して前記架橋された化合物を10重量部以上1000重量部以下の範囲内で含むことを特徴とする請求項28記載の電解質。

【請求項31】 前記シロキサン誘導体の平均分子量は100000以下であることを特徴とする請求項28記載の電解質。

【請求項32】 前記電解質塩は、リチウム塩であることを特徴とする請求項17記載の電解質。

【請求項33】 エーテル結合および架橋可能な官能基を有する架橋可能な化合物と、高分子化合物と、電解質塩とを混合して、架橋可能な化合物を架橋させることを特徴とする電解質の製造方法。

【請求項34】 紫外線、電子線、エックス線、ガンマ線、マイクロ波もしくは高周波を照射することにより、または加熱することにより架橋可能な化合物を架橋させることを特徴とする請求項33記載の電解質の製造方法。

【請求項35】 エーテル結合および架橋可能な官能基

を有する架橋可能な化合物と、高分子化合物とを混合して、架橋可能な化合物を架橋させたのち、電解質塩を加えることを特徴とする電解質の製造方法。

【請求項36】 紫外線、電子線、エックス線、ガンマ線、マイクロ波もしくは高周波を照射することにより、または加熱することにより架橋可能な化合物を架橋させることを特徴とする請求項35記載の電解質の製造方法。

【請求項37】 正極および負極と共に電解質を備えた電池であって、前記電解質は、エーテル結合および架橋基を有する化合物が架橋基において架橋された化合物と、高分子化合物と、電解質塩とを含むことを特徴とする電池。

【請求項38】 前記架橋された化合物は、少なくとも1つの架橋基を有する1種または2種以上の化合物が架橋基において架橋された構造を有することを特徴とする請求項37記載の電池。

【請求項39】 前記架橋された化合物は、エーテル結合を含む自由末端鎖を有することを特徴とする請求項37記載の電解質。

【請求項40】 前記架橋された化合物は、エステル化合物が架橋基において架橋された構造を有することを特徴とする請求項37記載の電池。

【請求項41】 前記架橋された化合物は、モノエステル化合物、ジエステル化合物およびトリエステル化合物のうちの少なくとも1種が架橋基において架橋された構造を有することを特徴とする請求項40記載の電池。

【請求項42】 前記架橋された化合物は、下記の化13で表されるジエステル化合物が架橋基において架橋された構造を有することを特徴とする請求項41記載の電池。

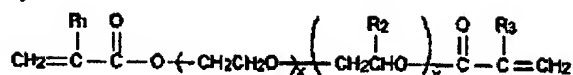
【化13】

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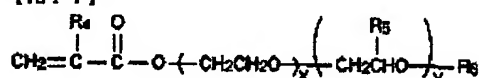
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(式中、 $\text{R}_1$ 、 $\text{R}_2$  および  $\text{R}_3$  は水素原子またはアルキル基を表し、 $x$  および  $y$  は  $x \geq 1$  かつ  $y \geq 0$  または  $x \geq 0$  かつ  $y \geq 1$  の整数である。)

【請求項43】 前記架橋された化合物は、更に、下記の化14で表されるモノエステル化合物が架橋基において架橋された構造を有することを特徴とする請求項42記載の電池。

【化14】

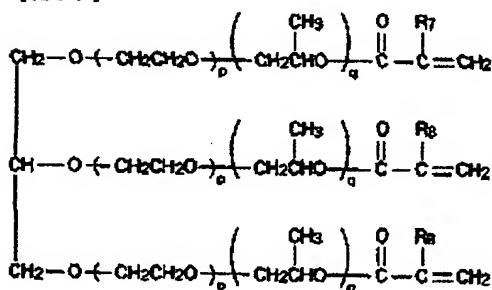


(式中、 $\text{R}_4$ 、 $\text{R}_5$  および  $\text{R}_6$  は水素原子またはアルキル基を表し、 $x$  および  $y$  は  $x \geq 1$  かつ  $y \geq 0$  または  $x \geq 0$  かつ  $y \geq 1$  の整数である。)

【請求項44】 前記架橋された化合物は、ジエステル化合物が架橋基において架橋された構造とモノエステル化合物が架橋基において架橋された構造とを、ジエステル化合物に対するモノエステル化合物の重量比(モノエステル化合物/ジエステル化合物)において0よりも大きく5.0以下の範囲内で有することを特徴とする請求項43記載の電池。

【請求項45】 前記架橋された化合物は、下記の化15で表されるトリエステル化合物が架橋基において架橋された構造を有することを特徴とする請求項41記載の電池。

【化15】

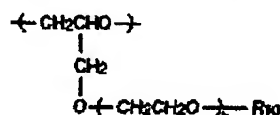


(式中、 $\text{R}_7$ 、 $\text{R}_8$  および  $\text{R}_9$  は水素原子またはアルキ

ル基を表し、 $p$  および  $q$  は  $p \geq 1$  かつ  $q \geq 0$  または  $p \geq 0$  かつ  $q \geq 1$  の整数である。)

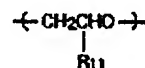
【請求項46】 前記架橋された化合物は、下記の化16で表される構成単位と下記の化17で表される構成単位とを含む共重合体が架橋基において架橋された構造を有することを特徴とする請求項37記載の電池。

10 【化16】



(式中、 $\text{R}_{10}$  はアルキル基、アルケニル基、シクロアルキル基、アリール基、アラルキル基およびテトラヒドロピラニル基のうちの少なくとも1種を表し、 $h$  は  $1 \leq h \leq 12$  の整数である。)

20 【化17】



(式中、 $\text{R}_{11}$  は活性水素基および不飽和二重結合を有する官能基のうちの少なくとも一方、または活性水素基および不飽和二重結合を有する官能基のうちの少なくとも一方並びに水素原子、アルキル基、アルケニル基、シクロアルキル基およびアリール基のうちの少なくとも1種を表す。)

30 【請求項47】 前記架橋された化合物に対する前記電解質塩の割合は、架橋された化合物におけるエーテル結合単位のモル数に対する電解質塩のモル数の割合(電解質塩のモル数/架橋された化合物におけるエーテル結合単位のモル数)で、0.0001以上5以下の範囲内であることを特徴とする請求項37記載の電池。

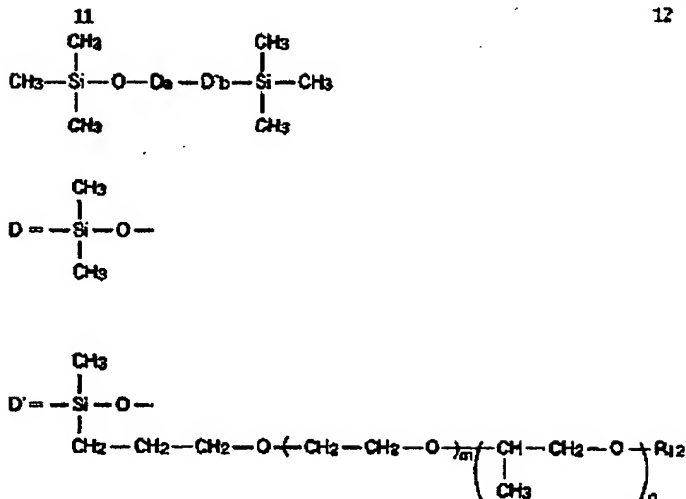
【請求項48】 前記高分子化合物は、下記の化18で表されるシロキサン誘導体を含むことを特徴とする請求項37記載の電池。

【化18】

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(式中、aは0から100の整数を表し、bは1から100の整数を表し、mは0から100の整数を表し、nは0から100の整数を表し、R<sub>12</sub>は水素原子またはアルキル基を表す。なお、b>1のときb個のD'は同じであっても異なってもよい。また、D'およびR<sub>12</sub>に含まれる水素原子はハロゲン原子で置き換えられていてもよい。)

【請求項49】 前記シロキサン誘導体に対する前記電解質塩の割合は、シロキサン誘導体におけるエーテル結合単位の数に対する電解質塩のモル数の割合（電解質塩のモル数/シロキサン誘導体におけるエーテル結合単位の数）で、0.0001以上5以下の範囲内であることを特徴とする請求項48記載の電池。

【請求項50】 前記シロキサン誘導体の100重量部に対して前記架橋された化合物を10重量部以上100重量部以下の範囲内で含むことを特徴とする請求項48記載の電池。

【請求項51】 前記シロキサン誘導体の平均分子量は10000以下であることを特徴とする請求項48記載の電池。

【請求項52】 前記電解質塩は、リチウム塩であることを特徴とする請求項37記載の電池。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】 本発明は、高分子化合物と電解質塩を含む電解質用組成物ならびに電解質およびその製造方法およびそれを用いた電池に関する。

【0002】

【従来の技術】 近年になって、カメラ一体型VTR（ビデオテーブルコード）、携帯電話あるいはラップトップコンピュータなどの携帯用電気製品が急速に普及しつつあり、このような電子機器のデバイスとして、電気化学デバイスの更なる高性能化が必要とされている。

【0003】 従来、二次電池などの電気化学デバイス

は、イオン伝導を司る物質として、水あるいは可燃性の有機溶媒などに電解質塩を溶解させた液状の電解質が用いられてきた。しかし、液状の電解質は漏液などの問題があり、このため金属製の容器を用い密封性を確保する必要があった。そのため、一般に、このような電気化学デバイスは重量が重く、また、密封工程に煩雑さが伴い、形状の自由度も低かった。そこで、イオン伝導性固体からなるいわゆる固体電解質を用いる研究が活発になされている。固体電解質には液漏れの心配が無く、そのための密封工程を簡略できると共に、デバイスを軽量化できる、更にポリマの優れたフィルム成型性により形状選択性の自由度が高いなどの利点がある。

【0004】 この固体電解質は、一般に、マトリクス高分子とイオン解離の可能な電解質塩とからなっている。このうち、マトリクス高分子は、イオン解離能を保持しており、このイオン伝導性固体を固体化する役目と電解質塩に対する溶媒としての役目といった双方の機能を持っている。このような固体電解質については、1978年に、グルノーブル大学（仏）のArmand氏によりポリエチレンオキサイドに過塩素酸リチウムを溶解させた系で1×10<sup>-2</sup> S/cm程度のイオン伝導度が得られたとの報告がされており、それ以来、ポリエーテル結合を有するポリマを中心に多岐にわたる高分子材料について検討が現在も高活発に行われている。

【0005】

【発明が解決しようとする課題】 このポリエチレンオキッドに代表されるような直鎖状ポリエーテルをマトリクスとして用いた固体電解質は、マトリクスポリマのガラス転移温度以上の無定形相中を、溶解しているイオンが高分子鎖の局所的セグメント運動で移動することにより、イオン伝導を示すようになっている。

【0006】 しかしながら、半結晶性のポリマであるポリエチレンオキッドのような直鎖状のマトリクス中に溶解しているイオン、特にカチオンは、高分子鎖との相互

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作用により強く配位し、これが疑似架橋点となって部分的な結晶化を引き起こし、セグメント運動が低下してしまうという問題があった。従って、室温下でより高いイオン伝導度を実現するためには、電解質塩の解離能が高く、また、マトリクス内でイオンが移動し易い極定形（アモルファス）領域が多く存在し、また、ポリマのガラス転移点を低く保つような分子設計を行うことが望まれている。

【0007】なお、この種の分子設計としては、例えば、ポリエチレンオキシド骨格に分岐構造を導入することによりイオン伝導度を向上させる試みが行われている（渡邊正義、Netsu Sokutei 24(1) pp12-21, 1996）。しかし、この種のポリマは、その合成方法が煩雑であるという問題がある。

【0008】また、他にも、マトリクス高分子に三次元網目構造を導入し、ポリマの結晶化を阻害させる試みもなされており、この種の分子設計の例として、ポリオキシアルキレン成分を有するアクリル系あるいはメタクリル系モノマを重合させる方法が報告されている（特開平5-25353号公報）。しかし、アルカリ金属塩のモノマへの溶解性が低いために十分なイオン伝導度が得られないといった問題がある。よって、これらに代わる新しい固体電解質が求められている。

【0009】本発明はかかる問題点に鑑みてなされたもので、その目的は、高いイオン伝導性を示すことができる電解質用組成物ならびに電解質およびその製造方法およびそれを用いた電池を提供することにある。

【0010】

【課題を解決するための手段】本発明による電解質用組成物は、エーテル結合および架橋可能な官能基を有する架橋可能な化合物と、高分子化合物と、電解質塩とを含むものである。

【0011】本発明による電解質は、エーテル結合および架橋基を有する化合物が架橋基において架橋された化合物と、高分子化合物と、電解質塩とを含むものである。

【0012】本発明による電解質の製造方法は、エーテル結合および架橋可能な官能基を有する架橋可能な化合物と、高分子化合物と、電解質塩とを混合して、架橋可能な化合物を架橋させるものである。

【0013】本発明による他の電解質の製造方法は、エーテル結合および架橋可能な官能基を有する架橋可能な化合物と、高分子化合物とを混合して、架橋可能な化合物を架橋させたのち、電解質塩を加えるものである。

【0014】本発明による電池は、正極および負極と共に電解質を備えたものであって、電解質は、エーテル結合および架橋基を有する化合物が架橋基において架橋された化合物と、高分子化合物と、電解質塩とを含むものである。

【0015】本発明による電解質用組成物では、エーテ

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ル結合および架橋可能な官能基を有する架橋可能な化合物と、高分子化合物とを含んでいるので、高いイオン伝導性を示す電解質が得られる。

【0016】本発明による電解質では、エーテル結合および架橋基を有する化合物が架橋基において架橋された化合物の間に、高分子化合物と解離された電解質塩とが存在している。よって、高いイオン伝導性を示す。

【0017】本発明による電解質の製造方法では、エーテル結合および架橋可能な官能基を有する架橋可能な化合物と、高分子化合物と、電解質塩とを混合されたのち、架橋可能な化合物が架橋される。

【0018】本発明による電解質の製造方法では、エーテル結合および架橋可能な官能基を有する架橋可能な化合物と、高分子化合物とを混合され、架橋可能な化合物が重合されたのち、電解質塩が加えられる。

【0019】本発明による電池では、電解質塩の解離により生じたイオンが負極と正極との間で電解質中を移動することにより放電する。ここでは、本発明の電解質を備えているので、高いイオン伝導性を有しており、優れた電池性能を示す。

【0020】

【発明の実施の形態】以下、本発明の実施の形態について詳細に説明する。

【0021】本発明の一実施の形態に係る電解質は、エーテル結合および架橋基を有する化合物が架橋基において架橋された三次元網目構造を有する化合物と、高分子化合物と、電解質塩とを含んでいる。このように三次元網目構造を有する架橋された化合物を含んでいるのは、高分子化合物のみでは分子量が低くなるにしたがって流動性を示すので成膜をすることができず、また高分子量体においても十分な膜強度を得にくいからである。すなわち、この三次元網目構造を有する架橋された化合物は、高分子化合物と電解質塩とをその三次元網目構造の間に保持し、成膜性と強い膜強度を担保するためのものである。

【0022】この架橋された化合物としては、例えば、直鎖状の化合物および樹状の化合物の少なくとも一方が架橋基において架橋された構造を有するものが挙げられ、側鎖を有していてもよい。なお、この架橋された化合物は、例えば、少なくとも一部に鎖状の原子配列を含む化合物が架橋された構造を有していればよく、鎖状化合物が架橋されたものに限られず、一部に環状の原子配列を有する化合物が架橋された構造を有していてもよい。すなわち、この架橋された化合物は、主鎖または側鎖に環状の原子配列を有していてもよい。

【0023】また、この架橋された化合物は、架橋基を少なくとも1つ有する化合物が架橋されたものであればよく、どのような位置に架橋基を有する化合物が架橋されたものであってもよい。例えば、直鎖状の化合物が架橋されたもの場合には、両端に架橋基を有する化合物

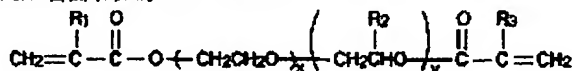
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が架橋されていてもよく、片端に架橋基を有する化合物が架橋されていてもよい。樹状の化合物が架橋されたもの場合には、少なくとも1つの末端に架橋基を有する化合物が架橋されていてもよい。すなわち、この架橋された化合物は、高分子化合物および電解質塩を保持することができる三次元網目構造を形成していればよい。

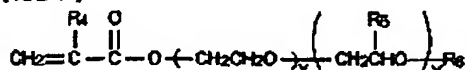
【0024】但し、より高いイオン伝導度を実現するには、架橋基が配されずかつエーテル結合を含む自由末端を少なくとも1つ有する化合物が架橋された構造を有することが好ましい。これにより三次元網目構造に分岐構造が導入され、エーテル結合を含む自由末端側鎖を有することになるからである。例えば、直鎖状の化合物が架橋された化合物により構成される場合には、両端に架橋基を有する化合物が架橋された構造に加えて、片端に架橋基を有する化合物が架橋された構造を有することが好ましい。また、樹状の化合物が架橋された化合物により構成される場合には、架橋基が配されない自由末端を少なくとも1つ有する樹状の化合物が架橋された構造を有することが好ましい。更に、直鎖状の化合物と樹状の化合物とが架橋された化合物により構成される場合には、片端に架橋基を有する直鎖状の化合物が架橋された構造、または架橋基が配されない自由末端を少なくとも1\*



式中、 $\text{R}_1$ 、 $\text{R}_2$  および  $\text{R}_3$  は水素原子または炭素数1～6のアルキル基を表し、それぞれは同一であっても異なっているてもよい。また、 $x$  および  $y$  は  $x \geq 1$  かつ  $y \geq 0$  または  $x \geq 0$  かつ  $y \geq 1$  の整数である。

【0027】この場合、例えば、化19に示した一般式で表されるジエステル化合物が架橋基において架橋された構造に加えて、化20に示した一般式で表されるオキシアルキレン構造を含むモノエステル化合物が架橋基において架橋された構造を有する方が好ましい。上述したように、エーテル結合を含む自由末端側鎖が導入され、より高いイオン伝導度を得ることができるからである。

【化20】



式中、 $\text{R}_4$ 、 $\text{R}_5$  および  $\text{R}_6$  は水素原子または炭素数1～6のアルキル基を表し、それぞれは同一であっても異なっているてもよい。また、 $x$  および  $y$  は  $x \geq 1$  かつ  $y \geq 0$  または  $x \geq 0$  かつ  $y \geq 1$  の整数である。

【0028】その場合、ジエステル化合物が架橋基において架橋された構造とモノエステル化合物が架橋基において架橋された構造とは、ジエステル化合物に対するモノエステル化合物の重量比（モノエステル化合物/ジエステル化合物）において0よりも大きく5.0以下の範囲内であることが好ましい。ジエステル化合物が架橋さ

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\*つ有する樹状の化合物が架橋された構造を有することが好ましい。

【0025】この架橋された化合物は、また、エーテル結合、エステル結合あるいはウレタン結合などのどのような結合による架橋構造を有していてもよい。例えば、アリル基、ビニル基、アクリレート基、メタクリレート基、シクロオレフィン構造を有する官能基あるいは活性水素基などを有する化合物がそれらにおいて架橋された架橋構造を有するものでもよい。また、複数の異なる架橋構造を1化合物内に有していてもよい。

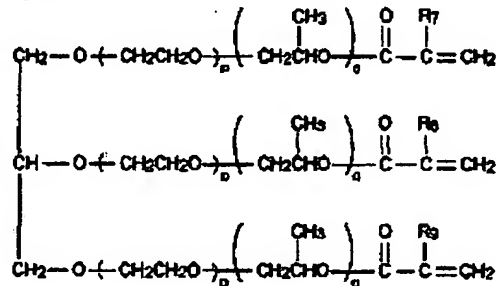
【0026】このような架橋された化合物としては、具体的には、エステル化合物が架橋基において架橋された架橋構造を有するものが挙げられ、中でも、モノエステル化合物、ジエステル化合物およびトリエステル化合物のうちの少なくとも1種が架橋された構造を有するものが好ましい。例えば、ジエステル化合物が架橋された構造を有する化合物としては、化19に示した一般式で表されるオキシアルキレン構造を含むジエステル化合物が架橋基において架橋された構造を有するものが挙げられ

【化19】

れた構造が少ないと三次元網目構造を構成することができず、モノエステル化合物が架橋された構造が少ないとエーテル結合を含む自由末端側鎖が少なくなりイオン伝導度を高くすることができないからである。

【0029】また、例えば、トリエステル化合物が架橋された構造を有する化合物としては、化21に示した一般式で表されるオキシアルキレン構造を含むトリエステル化合物が架橋基において架橋された構造を有するものが挙げられる。

【化21】



式中、 $\text{R}_7$ 、 $\text{R}_8$  および  $\text{R}_9$  は水素原子または炭素数1～6のアルキル基を表し、それぞれは同一であっても異なっているてもよい。また、 $p$  および  $q$  は  $p \geq 1$  かつ  $q \geq 0$  または  $p \geq 0$  かつ  $q \geq 1$  の整数である。

【0030】この化21に示したトリエステル化合物の

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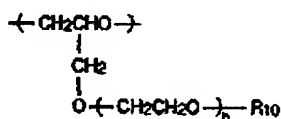
オキシエチレンユニットとオキシプロピレンユニットとの組成比は特に限定されるものではないが、 $0.1 \leq q/p \leq 4$ の範囲内であることが好ましい。オキシエチレンユニットが少なすぎると機械的強度が低下し、多すぎるともろくなるからである。また、オキシエチレンユニットとオキシプロピレンユニットとの結合様式はブロック状またはランダム状のいずれでも良いが、ランダム状の方が好ましい。

【0031】この場合も、上述したように、例えば、化21に示した一般式で表されるトリエステル化合物が架橋基において架橋された構造に加えて、化20に示した一般式で表されるモノエステル化合物が架橋基において架橋された構造を有する方が好ましい。

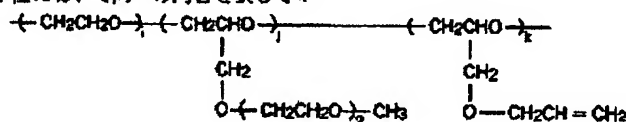
【0032】また、架橋された化合物としては、これらの他にも、例えば、化22に示した構成単位と化23に示した構成単位とを含む共重合体が架橋基において架橋された構造を有するものが挙げられる。この化合物によれば、化22に示した構成単位によりエーテル結合を含む自由末端側鎖が導入され、高いイオン伝導度を得ることができるので好ましい。

【0033】

【化22】



式中、 $R_{10}$ は炭素数1~12のアルキル基、炭素数2~18のアルケニル基、炭素数3~8のシクロアルキル基、炭素数6~18のアリール基、炭素数7~12のアラルキル基およびテトラヒドロピラニル基のうちの少なくとも1種を表し、 $h$ は $1 \leq h \leq 12$ の整数である。なお、 $R_{10}$ は全ての構成単位において同一の内容を表して\*



式中、 $i$ 、 $j$ および $k$ は1以上の整数である。

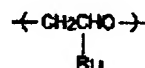
【0036】なお、この架橋された化合物には、これらの構造の異なる複数の化合物を混合して用いてもよい。例えば、化19に示したエステル化合物が架橋された構造を有する化合物と、化22に示した構成単位と化23に示した構成単位とを含む共重合体が架橋された構造を有する化合物とを混合して用いてもよい。

【0037】高分子化合物は、電解質塩を溶解するためのものである。この高分子化合物としては、例えば、ケイ素(Si)と酸素(O)との鎖状結合を基本骨格に持ち、ケイ素に側鎖Rが付加された(SiOCH<sub>2</sub>R)で表される鎖状型シロキサン誘導体が挙げられる。この

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\*いてもよく、構成単位により異なる内容を表していてもよい。

【化23】



式中、 $R_{11}$ は活性水素基および不飽和二重結合を有する官能基のうちの少なくとも一方、または活性水素基および不飽和二重結合を有する官能基のうちの少なくとも一方と水素原子、アルキル基、アルケニル基、シクロアルキル基およびアリール基のうちの少なくとも1種とを表す。なお、 $R_{11}$ は全ての構成単位において同一の内容を表していてもよく、構成単位により異なる内容を表していてもよい。

【0034】この共重合体における化22に示した構成単位と化23に示した構成単位との組成比は特に限定されるものではなく、化22に示した構成単位と化23に示した構成単位との結合様式はブロック状またはランダム状のいずれでもよい。ちなみに、この共重合体において架橋可能な官能基は活性水素基および不飽和二重結合を有する官能基である。不飽和二重結合を有する官能基としては、アリル基、ビニル基、アクリレート基、メタクリレート基あるいはシクロオレフィン構造を有する官能基などが挙げられる。

【0035】このような構造を有する架橋された化合物としては、具体的には、化24に示した一般式で表される共重合体が架橋基において架橋されたものが好ましい。化24に示した共重合体は、化22において $k$ が2で $R_{10}$ がメチル基よりなる構成単位と、化23において $R_{11}$ が水素原子よりなる構成単位と、化23において $R_{11}$ がアリル基よりなる構成単位とを含むものである。

【化24】

場合、置換基または側鎖基Rは一価の有機基であり、側鎖基Rが適度に変化することにより、電解質塩を溶解することができる構造となる。よって、このシロキサン誘導体の平均分子量は10000以下であることが好ましい。

【0038】また、このシロキサン誘導体は、電解質塩を有効に溶解し得ると共に、エーテル結合を有する化合物が架橋基において架橋された化合物との高い相溶性が必要とされるので、置換基または側鎖基Rにエーテル結合を含むことが好ましい。このような側鎖基Rとしてはエトキシ基、プロポキシ基、ブトキシ基、メトキシ基などのアルコキシ基がある。また、側鎖基R中の水素が水



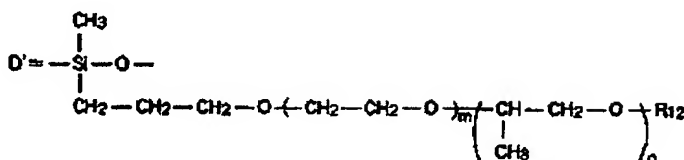
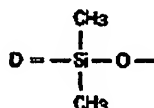
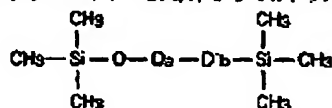
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ウ素またはフッ素などのハロゲン元素で置き換えられていてもよい。

【0039】このようなシロキサン誘導体としては、具\*



式中、aは0から100の整数を表し、bは1から100の整数を表し、mは0から100の整数を表し、nは0から100の整数を表し、R<sub>12</sub>は水素原子またはアルキル基を表す。なお、b>1のときb個のD'は同じであっても異なってもよい。また、D'およびR<sub>12</sub>に含まれる水素原子はハロゲン原子で置き換えられていてもよい。

【0040】電解質塩は、解離によりイオン伝導性を持たせるためのものであり、化26に示した一般式で表される。

【化26】AB

式中、Aはカチオン、Bはアニオンを表す。

【0041】この電解質塩としては軽金属塩などが挙げられ、具体的には、リチウム(Li)塩、ナトリウム(Na)塩あるいはカリウム(K)塩などのアルカリ金属塩、またはカルシウム(Ca)塩あるいはマグネシウム(Mg)塩などのアルカリ土金属塩などから目的に応じて用いられる。なお、リチウム塩としてはLiClO<sub>4</sub>、LiBF<sub>4</sub>、LiPF<sub>6</sub>、LiAsF<sub>6</sub>、LiAlCl<sub>4</sub>、LiSbF<sub>6</sub>、LiCF<sub>3</sub>SO<sub>3</sub>、LiN(CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>、LiC(F<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>、LiCF<sub>3</sub>CO<sub>2</sub>、LiN(CF<sub>3</sub>CO<sub>2</sub>)<sub>2</sub>などがあり、ナトリウム塩としてはNaClO<sub>4</sub>、NaBF<sub>4</sub>、NaSCNなどがあり、カリウム塩としてはKBF<sub>4</sub>などがある。これらは、必要に応じていずれか1種または2種以上が混合されて用いられる。

【0042】なお、架橋された化合物と高分子化合物との割合は、高分子化合物としてシロキサン誘導体を用いた場合、シロキサン誘導体の100重量部に対して架橋された化合物を10重量部以上10000重量部以下の範囲内とすることが好ましい。シロキサン誘導体の分子

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\* 体的には、化25に示した一般式で表されるものが好ましい。

【化25】

量または電解質の用途にもよるが、架橋された化合物が10重量部より少ないと膜強度が十分ではなく、1000重量部を超えると膜が脆くあるいは固くなるなどの不都合が生じるからである。

【0043】また、架橋された化合物に対する電解質塩の割合は、架橋された化合物におけるエーテル結合単位のモル数に対する電解質塩のモル数の割合（電解質塩のモル数／架橋された化合物におけるエーテル結合単位のモル数）で、0.0001以上5以下の範囲内であることが好ましい。シロキサン誘導体に対する電解質塩の割合も、同様に、シロキサン誘導体におけるエーテル結合単位のモル数に対する電解質塩のモル数の割合（電解質塩のモル数／シロキサン誘導体におけるエーテル結合単位のモル数）で、0.0001以上5以下の範囲内であることが好ましい。この範囲内において高いイオン伝導度を得ることができるからである。

【0044】ちなみに、化19に示したジエステル化合物1molに含まれるエーテル結合単位のモル数は(1+x+y)molであり、化24に示した共重合体1molに含まれるエーテル結合単位のモル数は{i+(1+1+2)×j+(1+1)×k}molである。

【0045】このような構成を有する電解質は、次のような電解質用組成物を用い、次のようにして製造することができる。

【0046】まず、エーテル結合および架橋可能な官能基を有する架橋可能な化合物と、高分子化合物と、電解質塩とを含む電解質用組成物を用意する。このうち高分子化合物および電解質塩は上述した通りである。例えば、高分子化合物としてはシロキサン誘導体を用意し、電解質塩としては少なくとも1種のリチウム塩を用意する。

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【0047】架橋可能な化合物は、架橋により上述した三次元網目構造を有する架橋された化合物となるものであり、直鎖状の化合物でも、筒状の化合物でも、それらの混合物でもよく、それらは側鎖を有していてもよい。また、少なくとも一部に鎖状の原子配列を有していれば鎖状化合物でも、主鎖または側鎖の一部に環状の原子配列を有する化合物でもよい。更に、架橋可能な官能基を少なくとも1つ有していれば数はいくつでもよく、その位置もどこでもよい。例えば、直鎖状の化合物の場合には、架橋可能な官能基を両端に有していてもよく、片端

【0048】また、架橋可能な化合物は架橋可能な官能基としてどのような官能基を有しているものでもよい。具体的には、エステル化合物あるいはアリル基を有する化合物などが挙げられる。例えば、エステル化合物であれば、モノエステル化合物、ジエステル化合物、トリエステル化合物あるいはエステル結合を4以上有しているものでもよい。更に、この架橋可能な化合物は架橋可能な官能基の数が異なるものを複数種含んでいてもよく、架橋可能な官能基の種類が異なるものを複数種含んでいてもよい。

【0049】このような架橋可能な化合物としては、例えば、化19に示した一般式で表されるジエステル化合物、化20に示した一般式で表されるモノエステル化合物および化21に示した一般式で表されるトリエステル化合物などが挙げられる。なお、化19に示したジエステル化合物を用いる場合には、この化合物に加えて、化20に示したモノエステル化合物を含む方が好ましい。これは、上述したように、架橋した際にエーテル結合を含む自由末端側鎖を形成するためである。この場合、ジエステル化合物に対するモノエステル化合物の重量比（モノエステル化合物/ジエステル化合物）は、0よりも大きく5.0以下の範囲内であることが好ましい。また、化21に示したトリエステル化合物を用いる場合にも、この化合物に加えて、化20に示したモノエステル化合物を含む方が好ましい。

【0050】他にも架橋可能な化合物としては、例えば、化22に示した構成単位と化23に示した構成単位とを含む共重合体などが挙げられる。具体的には、化24に示した共重合体が好ましい。

【0051】なお、架橋可能な化合物と高分子化合物との割合は、高分子化合物としてシロキサン誘導体を用いた場合、上述した通り、シロキサン誘導体の100重量部に対して架橋可能な化合物を10重量部以上1000重量部以下の範囲内とすることが好ましい。また、上述した通り、架橋可能な化合物に対する電解質塩の割合は、架橋可能な化合物におけるエーテル結合単位のモル数に対する電解質塩のモル数の割合（電解質のモル数/

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架橋可能な化合物におけるエーテル結合単位のモル数)で、0.0001以上5以下の範囲内であることが好ましく、シロキサン誘導体に対する前記電解質塩の割合は、シロキサン誘導体におけるエーテル結合単位のモル数に対する電解質塩のモル数の割合（電解質のモル数/シロキサン誘導体におけるエーテル結合単位のモル数）で、0.0001以上5以下の範囲内であることが好ましい。

【0052】次いで、これらエーテル結合および架橋可能な官能基を有する架橋可能な化合物と、高分子化合物と、電解質塩とを混合し、均一な混合物を作製する。なお、その際、必要に応じて重合開始剤も添加する。重合開始剤は重合を電子線の照射により行う場合以外には必要となり、光重合開始剤と熱重合開始剤とがある。

【0053】光重合開始剤としては、例えば、アセトフェノン、トリクロロアセトフェノン、2-ヒドロキシ-2-メチルプロピオフェノン、2-ヒドロキシ-2-メチルイソプロピオフェノン、1-ヒドロキシシクロヘキシルケトン、ベンゾイソエーテル、2,2-ジエトキシアセトフェノンあるいはベンジルジメチルケタールがある。また、熱重合開始剤としては、例えば、クメンヒドロパーオキシド、1-ブチルヒドロパーオキシド、ジクミルパーオキシドあるいはジ-1-ブチルパーオキシドなどの高温重合開始剤、または過酸化ベンゾイル、過酸化ラウロイル、過硫酸塩あるいはアゾビスイソブチロニトリルなどの開始剤、またはレドックス開始剤がある。重合開始剤としては、これらのうちの1種または2種以上を混合して用いてもよい。

【0054】なお、重合開始剤は、エーテル結合および架橋可能な官能基を有する架橋可能な化合物100重量部に対して0.1~1.0重量部の範囲で添加する。0.1重量部より少ないと重合速度が著しく低下してしまい、1.0重量部を超えて添加しても効果は変わらないからである。

【0055】続いて、この混合物に紫外線、電子線、エックス線、ガンマ線、マイクロ波もしくは高周波を照射するか、またはこの混合物を加熱し、架橋可能な化合物を重合させる。これにより、架橋可能な化合物が架橋され、本実施の形態に係る電解質が得られる。

【0056】なお、架橋可能な化合物と、高分子化合物と、電解質塩とを混合する際に、アセトニトリルなどの適宜な有機溶媒を用いて混合物を形成するようにしてもよい。この場合、架橋可能な化合物を重合させたのち、真空中または減圧下において放置または加熱するなどの乾燥処理を行い、有機溶媒を除去するようにしてもよく、また、その混合物を得たのち、乾燥処理を行い、有機溶媒を除去してから架橋可能な化合物を重合させるようにしてもよい。

【0057】また、この電解質は、次のようにしても製造することができる。

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【0058】まず、先の製造方法と同様に、エーテル結合および架橋可能な官能基を有する架橋可能な化合物と、高分子化合物と、電解質塩とを用意する。次いで、架橋可能な化合物と、高分子化合物とを混合する。その際、必要に応じて重合開始剤を混合する。続いて、先の製造方法と同様にして架橋可能な化合物を重合させ、架橋可能な化合物が架橋基において架橋された化合物と高分子化合物との混合物を形成する。そのうち、電解質塩を有機溶剤に溶解し、この混合物に浸透させ、有機溶剤を乾燥除去する。これにより、本実施の形態に係る電解質が得られる。

【0059】なお、重合反応の条件は特に限定されるものではないが、重合反応は装置の簡便性、コスト面から考えて、紫外線照射あるいは加熱重合が好ましい。また、例えば、アルカリ金属イオンを用いたリチウム電池、リチウムイオン電池、ナトリウム電池に用いる電解質を作製する場合には、電解質用組成物（すなわち、架橋可能な化合物、高分子化合物およびアルカリ金属塩）およびこれらを混合する際の溶媒および重合開始剤は十分に脱水処理を施すことが好ましく、製造中の雰囲気も低湿度とすることが好ましい。これは、アルカリ金属塩の種類によっては水分と反応することにより分解してしまうものがあるからである。また、電池として用いた場合に負極においてアルカリ金属と水分とが激しく反応してしまうからである。

【0060】このようにして製造される電解質は、次のようにして電池に用いられる。ここでは、リチウムを用いた二次電池の例を挙げ、図面を参照して、以下に説明する。

【0061】図1は、本実施の形態に係る電解質を用いた二次電池の断面構造を表すものである。なお、図1に示したものは、いわゆるペーパー型といわれるものである。この二次電池は、負極11と正極12とが本実施の形態に係る電解質13を介して積層されている。これらの側面には絶縁パッキン14が配設されており、負極11と正極12とを間に挟むように配設された一対の外装部材15、16の周縁部と接合されることにより密閉されている。

【0062】負極11は、例えば、リチウム金属、またはリチウムイオンを吸蔵および脱離することが可能な金属、合金あるいは炭素材料を含有している。この炭素材料は、所定の温度および雰囲気にて調製されたものであり、例えば、熱分解炭素類、石油コークスもしくはピッチコークスなどのコークス類、人造黒鉛類、天然黒鉛類、アセチレンブラックなどのカーボンブラック、ガラス状炭素類、有機高分子材料焼成体あるいは炭素繊維などが用いられている。なお、有機高分子材料焼成体というのは、有機高分子材料を不活性ガス雰囲気中または真空中において500℃以上の適当な温度で焼成したものである。

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【0063】正極12は、例えば、正極活性物質として、 $TiS_2$ 、 $MoS_2$ 、 $NbSe$ 、あるいはV、O、などのリチウムを含有しない金属硫化物もしくは酸化物、またはリチウムを含有するリチウム複合硫化物もしくはリチウム複合酸化物を含有している。特に、エネルギー密度を高くするには、 $Li_xMO_y$ を主体とするリチウム複合酸化物を含んでいることが好ましい。なお、Mは1種類以上の遷移金属が好ましく、具体的には、コバルト（Co）、ニッケル（Ni）およびマンガン（Mn）のうちの少なくとも1種が好ましい。また、xは、通常、 $0.05 \leq x \leq 1.10$ の範囲内の値である。このようなリチウム複合酸化物の具体例としては、 $LiCoO_2$ 、 $LiNiO_2$ 、 $Li_xNi_yCo_{1-x-y}O_2$ （但し、xおよびyの値は電池の充放電状態によって異なり、通常、 $0 < x < 1$ 、 $0.7 < y \leq 1$ である。）あるいは $LiMn_2O_4$ などが挙げられる。

【0064】なお、このリチウム複合酸化物は、例えば、リチウムの炭酸塩、硝酸塩、酸化物あるいは水酸化物と、遷移金属の炭酸塩、硝酸塩、酸化物あるいは水酸化物とを所望の組成に応じて粉砕混合し、酸素雰囲気中において600～1000℃の範囲内の温度で焼成することにより調製される。

【0065】電解質13は、ここでは電解質塩としてリチウム塩を含んでいる。また、電解質13は、この二次電池ではセパレータとしての役目も兼ねている。すなわち、負極11と正極12とを隔離し、両極の接触による電流の短絡を防止しつつリチウムイオンを通過させるようになっている。なお、必要に応じて、図示しないセパレータを負極11と電解質13との間、正極12と電解質13の間あるいは電解質13の中に備えるようにしてもよい。セパレータとしては、ポリテトラフルオロエチレン、ポリプロピレンあるいはポリエチレンなどの合成樹脂製の不織布からなるもの、セラミックフィルムからなるもの、または多孔質薄膜フィルムからなるものなどがある。

【0066】このような構成を有する二次電池は次のように作用する。

【0067】この二次電池では、充電を行うと、例えば、正極12からリチウムがイオンとなって脱離し、電解質13を介して負極11に吸蔵される。放電を行うと、例えば、負極11からリチウムがイオンとなって脱離し、電解質13を介して正極12に戻り吸蔵される。ここで、電解質13は、エーテル結合および架橋基を有する化合物が架橋基において架橋された化合物を含んでおり、その三次元網目構造の間に高分子化合物とリチウムイオンとを保持している。よって、成膜性と強い膜強度とを有すると共に、高いイオン伝導性も有している。従って、優れた電池性能を有する。

【0068】このように、本実施の形態に係る電解質によれば、エーテル結合および架橋基を有する化合物が架

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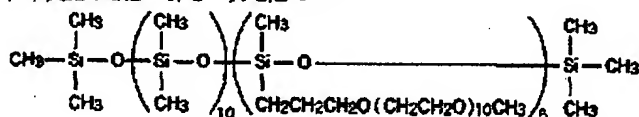
橋基において架橋された化合物と高分子化合物とを含むようにしたので、その三次元網目構造の間に高分子化合物と電解質塩とを保持することができる。よって、高い成膜性および強い膜強度を保持しつつ、イオン伝導性を高くすることができる。従って、この電解質を用いて電気化学デバイスを構成すれば、容易に高い性能を有する電気化学デバイスを得ることができる。

【0069】また、架橋された化合物がエーテル結合を含む自由末端側鎖を有するようにすれば、より高いイオン伝導度を得ることができる。

【0070】更に、本実施の形態に係る電解質用組成物によれば、エーテル結合および架橋可能な官能基を有する架橋可能な化合物と高分子化合物とを含むようにしたので、紫外線の照射または加熱によりこの架橋可能な化合物を容易に重合させることができる。よって、容易に本実施の形態に係る電解質を得ることができる。

【0071】加えて、本実施の形態に係る電解質の製造方法によれば、エーテル結合および架橋可能な官能基を有する架橋可能な化合物と高分子化合物とを混合したのち、架橋可能な化合物を重合させるようにしたので、容易に本実施の形態に係る電解質を得ることができる。

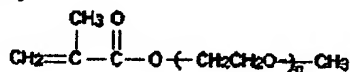
【0072】更にまた、本実施の形態に係る二次電池に\*



【化28】



【化29】



【0076】続いて、この混合溶液を減圧下において25℃に保ち、15重量部になるまでアセトニトリルを除去したのち、重合開始剤として0.05重量部の2,2-ジメトキシ-2,2'-フェニルアセトフェノンを添

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\*よれば、エーテル結合および架橋基を有する化合物が架橋基において架橋された化合物を含む電解質を用いるようにしたので、高い成膜性および強い膜強度を得ることができると共に、高いイオン伝導性も得ることができる。よって、容易に製造することができ、かつ高い性能を得ることができる。

【0073】

【実施例】更に、本発明の具体的な実施例について詳細に説明する。

10 【0074】（実施例1）まず、溶媒として50重量部のアセトニトリル（CH<sub>3</sub>CN）に、化27に示した1重量部のシロキサン誘導体と、このシロキサン誘導体に対して1.0mol/kgのリチウムビス（トリフルオロメチルスルホニル）イミド（Li（CF<sub>3</sub>SO<sub>2</sub>）<sub>2</sub>N））を加えて溶解した。次いで、これに架橋可能な化合物として化28に示した2重量部のポリエチレングリコールジメタクリレートと、化29に示した2重量部のメトキシポリエチレングリコールモノメタクリレートとを加え、均一な溶液になるまで混合した。

20 【0075】

【化27】

加して溶解させ、テフロン製の基板上に均一に塗布した。基板上に塗布したのち、25℃の空気中において紫外線を6mW/cm<sup>2</sup>の光量で20分間照射し、重合反応を行った。そののち、60℃の減圧下において6時間乾燥させ、厚さ100μmの電解質を得た。

30 【0077】この電解質の膜を面積0.7854cm<sup>2</sup>の円盤状に切り抜き、一對のステンレス製の電極に挟み込んで、交流インピーダンス法により25℃におけるイオン伝導度を求めた。その結果を表1に示す。

【0078】

【表1】

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	架橋可能な化合物の添加量 (重量部)			イオン 伝導度 (mS・cm)
	化28に示したジメタクリレート化合物	化29に示したモノメタクリレート化合物	化30に示したトリアクリレート化合物	
実施例1	2	2	—	0.15
2	0.5	0.5	—	0.22
3	0.125	0.125	—	0.43
4	1.33	2.66	—	0.23
5	0.83	0.166	—	0.51
6	—	—	4	0.016
7	—	—	1	0.067
8	—	—	0.25	0.31

【0079】(実施例2) 化28に示したポリエチレングリコールジメタクリレートの添加量を0.5重量部とし、化29に示したメトキシポリエチレングリコールモノメタクリレートの添加量を0.5重量部としたことを除き、実施例1と同様にして電解質を作製した。この電解質についても実施例1と同様にしてイオン伝導度を調べた。その結果を同様に表1に示す。

【0080】(実施例3) 化28に示したポリエチレングリコールジメタクリレートの添加量を0.125重量部とし、化29に示したメトキシポリエチレングリコールモノメタクリレートの添加量を0.125重量部としたことを除き、実施例1と同様にして電解質を作製した。この電解質についても実施例1と同様にしてイオン伝導度を調べた。その結果を同様に表1に示す。

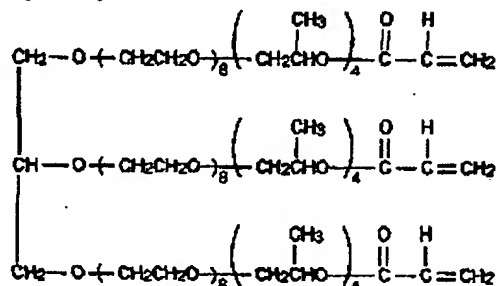
【0081】(実施例4) 化28に示したポリエチレングリコールジメタクリレートの添加量を1.33重量部とし、化29に示したメトキシポリエチレングリコールモノメタクリレートの添加量を2.66重量部としたことを除き、実施例1と同様にして電解質を作製した。この電解質についても実施例1と同様にしてイオン伝導度を調べた。その結果を同様に表1に示す。

【0082】(実施例5) 化28に示したポリエチレングリコールジメタクリレートの添加量を0.83重量部とし、化29に示したメトキシポリエチレングリコールモノメタクリレートの添加量を0.166重量部としたことを除き、実施例1と同様にして電解質を作製した。この電解質についても実施例1と同様にしてイオン伝導度を調べた。その結果を同様に表1に示す。

【0083】(実施例6) 化28に示したポリエチレングリコールジメタクリレートおよび化29に示したメトキシポリエチレングリコールモノメタクリレートに代えて、架橋可能な化合物として化30に示したトリアクリレート化合物を4重量部加えたことを除き、実施例1と同様にして電解質を作成した。この電解質についても実施例1と同様にしてイオン伝導度を調べた。その結果を同様に表1に示す。

【0084】

【化30】



【0085】(実施例7) 化28に示したポリエチレングリコールジメタクリレートおよび化29に示したメトキシポリエチレングリコールモノメタクリレートに代えて、化30に示したトリアクリレート化合物を1重量部加えたことを除き、実施例1と同様にして電解質を作成した。この電解質についても実施例1と同様にしてイオン伝導度を調べた。その結果を同様に表1に示す。

【0086】(実施例8) 化28に示したポリエチレングリコールジメタクリレートおよび化29に示したメトキシポリエチレングリコールモノメタクリレートに代えて、化30に示したトリアクリレート化合物を0.25重量部加えたことを除き、実施例1と同様にして電解質を作成した。この電解質についても実施例1と同様にしてイオン伝導度を調べた。その結果を同様に表1に示す。

【0087】(実施例9) まず、化27に示したシロキサン誘導体とリチウムビスイミドとを、シロキサン誘導体におけるエーテル結合単位のモル数に対するリチウムビスイミドのモル数の割合(リチウム塩のモル数/シロキサン誘導体におけるエーテル結合単位のモル数)が0.06となるように混合し、シロキサン誘導体の混合体を調整した。

【0088】次いで、化31に示した構成単位20.6 mol%と、化32に示した構成単位77.5 mol%

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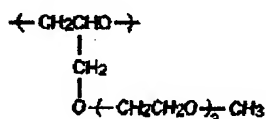
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と、化33に示した構成単位1.9mol%とからなる数平均分子量が82万の固体状ランダム共重合体を架橋可能な化合物として用意し、この共重合体とリチウムビスイミドとを、共重合体におけるエーテル結合単位のモル数に対するリチウムビスイミドのモル数の割合（リチウム塩のモル数/共重合体におけるエーテル結合単位のモル数）が0.06となるように混合して、共重合体の混合体を調整した。

【0089】

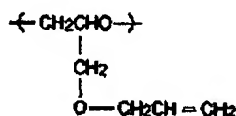
【化31】



【化32】



【化33】



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	混合比 (重量比) シロキサン誘導体の混合体/共重合体の混合体	イオン 伝導度 (mS/cm)
実施例9	5/5	0.166
10	2.5/7.5	0.385
11	7.5/2.5	0.0958
比較例	0/1	0.0103

【0093】（実施例10）シロキサン誘導体の混合体と共重合体の混合体とを、共重合体の混合体に対するシロキサン誘導体の混合体の重量比（シロキサン誘導体の混合体/共重合体の混合体）が7.5/2.5となるように混合したことを除き、実施例9と同様にして電解質を作製した。この電解質についても実施例1と同様にしてイオン伝導度を調べた。その結果を表2に示す。

【0094】（実施例11）シロキサン誘導体の混合体と共重合体の混合体とを、共重合体の混合体に対するシロキサン誘導体の混合体の重量比（シロキサン誘導体の混合体/共重合体の混合体）が2.5/7.5となるように混合したことを除き、実施例9と同様にして電解質を作製した。この電解質についても実施例1と同様にしてイオン伝導度を調べた。その結果を表2に示す。

【0095】（比較例）実施例9～11に対する比較例として、シロキサン誘導体の混合体を混合せず共重合体の混合体のみを用いたことを除き、実施例9と同様にして電解質を作製した。この電解質についても実施例1と同様にしてイオン伝導度を調べた。その結果を表2に示す。

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\*【0090】続いて、シロキサン誘導体の混合体と共重合体の混合体とを、共重合体の混合体に対するシロキサン誘導体の混合体の重量比（シロキサン誘導体の混合体/共重合体の混合体）が5/5となるようにアセトニトリル中において混合し、重合開始剤として2,2-ジメトキシ-2,2'-フェニルアセトフェノンを添加して混合溶液とした。

【0091】混合溶液を調整したのち、この混合溶液をテフロン製の基板上に均一に塗布し、25℃で減圧乾燥させ、25℃の空気中において紫外線を6mW/cm<sup>2</sup>の光量で20分間照射し、重合反応を行った。そののち、60℃の減圧下において6時間乾燥させ、厚さ100μmの電解質を得た。この電解質についても実施例1と同様にしてイオン伝導度を調べた。その結果を表2に示す。

【0092】

【表2】

【0096】実施例1～8および実施例9～11の結果から、実施例の電解質はいずれも1×10<sup>-3</sup>S/cmを超えるイオン伝導度が得られ、電池に使用するに十分な伝導性を有することが分かる。また、実施例9～11および比較例の結果から、実施例の電解質は比較例に比べて高いイオン伝導度が得られることも分かる。すなわち、エーテル結合および架橋基を有する化合物が架橋基において架橋された化合物と、シロキサン誘導体などの高分子化合物とを含むことにより、イオン伝導度を向上させることができ、この電解質を用いれば、優れた性能を有する電池が得られることが分かった。

【0097】以上、実施の形態および各実施例を挙げて本発明を説明したが、本発明は上記実施の形態および各実施例に限定されるものではなく、種々変形可能である。例えば、上記実施の形態においては、エーテル結合および架橋可能な官能基を有する架橋可能な化合物を互いに架橋させた場合について説明したが、エーテル結合を有する架橋可能な化合物と他の化合物とを架橋させるようにしてもよい。すなわち、エーテル結合および架橋基を有する化合物が架橋基において架橋された化合物と

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いうのは、エーテル結合および架橋基を有する化合物が架橋された構造を少なくとも一部に有していればよい。

【0098】また、上記実施の形態においては、エーテル結合および架橋可能な官能基を有する化合物として、化19、化20および化21に示したエステル化合物、および化22に示した構成単位と化23に示した構成単位とを含む共重合体を具体的に説明したが、エーテル結合および架橋可能な官能基を有していれば他の構造を有するものも同様に用いることができる。

【0099】更に、上記実施の形態および各実施例においては、高分子化合物としてシロキサン誘導体を説明したが、電解質塩を溶解することができる他の高分子化合物も同様に用いることができる。その際、複数の高分子化合物を用いるようにしてもよく、シロキサン誘導体に代えてまたはシロキサン誘導体と共に他の高分子化合物を用いてもよい。

【0100】加えて、上記実施の形態においては、リチウムを用いた二次電池について説明し、上記各実施例においては、電解質塩としてリチウム塩を用いた場合について説明したが、本発明は、ナトリウム塩あるいはカルシウム塩など他の電解質塩を用いた電解質および二次電池についても同様に適用することができる。

【0101】更にまた、上記実施の形態においては、ベーパー型の二次電池について説明したが、本発明は、ボタン型、コイン型、角型あるいはスパイラル構造を有する筒型など他の形状のものについても同様に適用することができる。

【0102】加えてまた、上記実施の形態においては、本発明の電解質を二次電池に用いる場合について説明したが、本発明の電解質および電解質用組成物は、一次電池などの他の電池についても用いることができる。更に、コンデンサ、キャパシタあるいはエレクトロクロミック素子などの他の電気化学デバイスに用いることもできる。なお、例えば、コンデンサなどに本発明の電解質を用いる場合には、電解質塩としてアンモニウム塩などの塩基性基塩を用いることもできる。

【0103】

【発明の効果】以上説明したように請求項1ないし請求項16のいずれか1項に記載の電解質用組成物によれば、エーテル結合および架橋可能な官能基を有する架橋可能な化合物と高分子化合物とを含むようにしたので、重合により容易に本発明の電解質を得ることができる。すなわち、高い成膜性および高いイオン伝導性を有する電解質を容易に得ることができるという効果を奏する。

【0104】また、請求項3、請求項7、請求項8ある

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いは請求項10に記載の電解質用組成物によれば、架橋可能な化合物として、架橋可能な官能基が配されずかつエーテル結合を含む自由末端を少なくとも1つ有する化合物を含むようにしたので、重合によりエーテル結合を含む自由末端側鎖を形成することができ、より高いイオン伝導度を有する電解質を得ることができるという効果を奏する。

【0105】更に、請求項17ないし請求項32のいずれか1項に記載の電解質によれば、エーテル結合および架橋基を有する化合物が架橋基において架橋された化合物と高分子化合物とを含むようにしたので、その三次元網目構造の間に高分子化合物と電解質塩とを保持することができる。よって、高い成膜性および強い膜強度を保持しつつ、イオン伝導性を高くすることができる。従って、この電解質を用いて電気化学デバイスを構成すれば、容易に高い性能を有する電気化学デバイスを得ることができるという効果を奏する。

【0106】加えて、請求項19記載の電解質によれば、架橋された化合物がエーテル結合を含む自由末端側鎖を有するようにしたので、また、請求項23または請求項24に記載の電解質によれば、化8で表されるモノエステル化合物が架橋された構造を有するようにしたので、また、請求項26に記載の電解質によれば、化10に示した構成単位を含む共重合体が架橋基において架橋された構造を有するようにしたので、より高いイオン伝導度を得ることができるという効果を奏する。

【0107】更にまた、請求項33ないし請求項36のいずれか1項に記載の電解質の製造方法によれば、エーテル結合および架橋可能な官能基を有する架橋可能な化合物と高分子化合物とを混合したのち、架橋可能な化合物を重合させるようにしたので、容易に本発明の電解質を得ることができるという効果を奏する。

【0108】加えてまた、請求項37ないし請求項52のいずれか1項に記載の電池によれば、本発明の電解質を用いるようにしたので、高い成膜性および強い膜強度を得ることができると共に、高いイオン伝導性も得ることができる。よって、容易に製造することができ、かつ高い性能を得ることができるという効果を奏する。

【図面の簡単な説明】

【図1】本発明の一実施の形態に係る電解質を用いた二次電池の構成を表す断面図である。

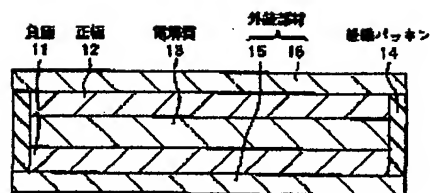
【符号の説明】

11…負極、12…正極、13…電解質、14…絶縁パッキング、15、16…外装部材

(18)

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【図1】



フロントページの続き

(51)Int.Cl.	識別記号	F I	ターマコード (参考)
H 0 1 G	9/025	H 0 1 M	6/18 E
	9/028		10/40 B
H 0 1 M	6/18	H 0 1 G	9/00 3 0 1 G
	10/40		9/02 3 3 1 G



## \* NOTICES \*

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3. In the drawings, any words are not translated.

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**CLAIMS**


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**[Claim(s)]**

[Claim 1] The constituent for electrolytes characterized by including the compound which has the functional group in which ether linkage and bridge formation are possible, and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt.

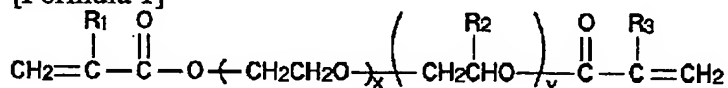
[Claim 2] The compound in which the aforementioned bridge formation is possible is a constituent for electrolytes according to claim 1 characterized by including one sort which has at least one functional group which can construct a bridge, or two sorts or more of compounds.

[Claim 3] The compound in which the aforementioned bridge formation is possible is a constituent for electrolytes according to claim 1 characterized by including the compound which has at least one free end which the functional group which can construct a bridge is not allotted and includes ether linkage.

[Claim 4] The compound in which the aforementioned bridge formation is possible is a constituent for electrolytes according to claim 1 characterized by including an ester compound.

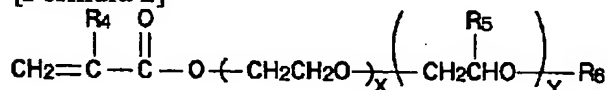
[Claim 5] The compound in which the aforementioned bridge formation is possible is a constituent for electrolytes according to claim 4 characterized by including at least one sort in a monoester compound, a diester compound, and a triester compound.

[Claim 6] The compound in which the aforementioned bridge formation is possible is a constituent for electrolytes according to claim 5 characterized by including the diester compound expressed with following \*\* 1.

**[Formula 1]**

(R1, R2, and R3 express a hydrogen atom or an alkyl group among a formula, and x and y are the integers of  $x \geq 1$  and  $y \geq 0$  or  $x \geq 0$ , and  $y \geq 1$ .)

[Claim 7] The compound in which the aforementioned bridge formation is possible is a constituent for electrolytes according to claim 6 characterized by including the monoester compound expressed with \*\* 2 of further the following.

**[Formula 2]**

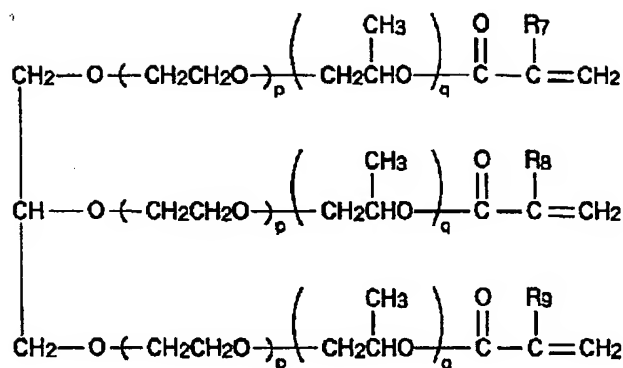
(R4, R5, and R6 express a hydrogen atom or an alkyl group among a formula, and X and Y are the integers of  $X \geq 1$  and  $Y \geq 0$  or  $X \geq 0$ , and  $Y \geq 1$ .)

[Claim 8] The weight ratio (a monoester compound / diester compound) of the aforementioned monoester compound to the aforementioned diester compound is a constituent for electrolytes according to claim 7 characterized by being within the limits of 5.0 or less more greatly than 0.

[Claim 9] The compound in which the aforementioned bridge formation is possible is a constituent for electrolytes according to claim 5 characterized by including the triester compound expressed with following \*\* 3.

**[Formula 3]**

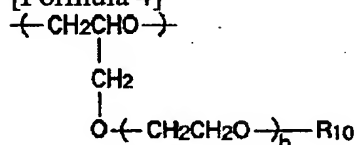
.../tran\_web.cgi\_ejje?u=http%3A%2F%2Fwww6.ipdl.jpo.go.jp%2FTokujitu%2Ftjitemcnt.ipdl%6/4/2003



(R7, R8, and R9 express a hydrogen atom or an alkyl group among a formula, and p and q are the integers of  $p \geq 1$  and  $q \geq 0$  or  $p \geq 0$ , and  $q \geq 1$ .)

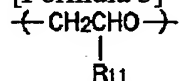
[Claim 10] The compound in which the aforementioned bridge formation is possible is a constituent for electrolytes according to claim 1 characterized by being a copolymer including the composition unit expressed with following \*\* 4, and the composition unit expressed with following \*\* 5.

[Formula 4]



(R10 expresses at least one sort in an alkyl group, an ARUKENIRU machine, a cycloalkyl machine, an aryl group, an aralkyl machine, and a tetrahydropyranyl group among a formula, and h is the integer of  $1 \leq h \leq 12$ .)

[Formula 5]

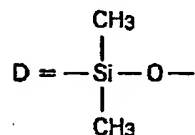
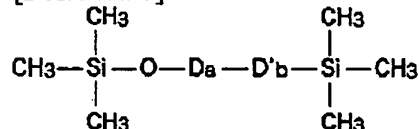


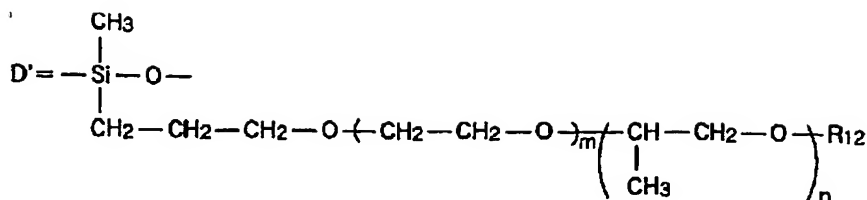
(the inside of the functional group in which R11 has an active hydrogen machine and an unsaturation double bond among a formula -- at least -- on the other hand -- or the inside of the functional group which has an active hydrogen machine and an unsaturation double bond -- on the other hand, at least one sort in a hydrogen atom, an alkyl group, an ARUKENIRU machine, a cycloalkyl machine, and an aryl group is expressed to a list at least)

[Claim 11] The rate of the aforementioned electrolyte salt to the compound in which the aforementioned bridge formation is possible is a constituent for electrolytes according to claim 1 characterized by being the rate (the number of mols of the ether linkage unit in the compound in which the number of mols / bridge formation of an electrolyte salt are possible) of the number of mols of an electrolyte salt to the number of mols of the ether linkage unit in the compound which can construct a bridge, and being within the limits of five or less [ 0.0001 or more ].

[Claim 12] The aforementioned high molecular compound is a constituent for electrolytes according to claim 1 characterized by including the siloxane derivative expressed with following \*\* 6.

[Formula 6]





(a expresses the integer of 0 to 100 among a formula, b expresses the integer of 1 to 100, m expresses the integer of 0 to 100, n expresses the integer of 0 to 100, and R<sub>12</sub> expresses a hydrogen atom or an alkyl group.) In addition, at the time of b > 1, even if b D' is the same, they may differ. Moreover, the hydrogen atom contained in D' and R<sub>12</sub> may be replaced by the halogen atom.

[Claim 13] The rate of the aforementioned electrolyte salt to the aforementioned siloxane derivative is a constituent for electrolytes according to claim 12 characterized by being the rate (the number of mols of the ether linkage unit in the number of mols / siloxane derivative of an electrolyte salt) of the number of mols of an electrolyte salt to the number of mols of the ether linkage unit in a siloxane derivative, and being within the limits of five or less [ 0.0001 or more ].

[Claim 14] The constituent for electrolytes according to claim 12 characterized by including the compound in which the aforementioned bridge formation is possible by within the limits below the 10000 weight sections more than 10 weight sections to the 100 weight sections of the aforementioned siloxane derivative.

[Claim 15] The average molecular weight of the aforementioned siloxane derivative is a constituent for electrolytes according to claim 12 characterized by being 100000 or less.

[Claim 16] The aforementioned electrolyte salt is a constituent for electrolytes according to claim 1 characterized by being lithium salt.

[Claim 17] The electrolyte characterized by the compound which has ether linkage and a bridge formation machine containing the compound over which the bridge was constructed in the bridge formation machine, a high molecular compound, and an electrolyte salt.

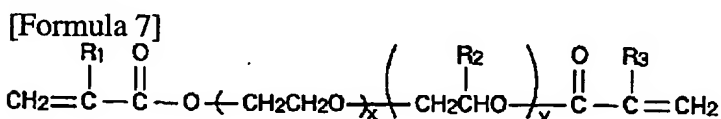
[Claim 18] The compound by which bridge formation was carried out [ aforementioned ] is an electrolyte according to claim 17 with which one sort which has at least one bridge formation machine, or two sorts or more of compounds are characterized by having the structure where the bridge was constructed in the bridge formation machine.

[Claim 19] The compound by which bridge formation was carried out [ aforementioned ] is an electrolyte according to claim 17 characterized by having a free end side chain including ether linkage.

[Claim 20] The compound by which bridge formation was carried out [ aforementioned ] is an electrolyte according to claim 17 with which an ester compound is characterized by having the structure where the bridge was constructed in the bridge formation machine.

[Claim 21] The compound by which bridge formation was carried out [ aforementioned ] is an electrolyte according to claim 20 characterized by having the structure where the bridge was constructed over at least one sort in a monoester compound, a diester compound, and a triester compound in the bridge formation machine.

[Claim 22] The compound by which bridge formation was carried out [ aforementioned ] is an electrolyte according to claim 21 with which the diester compound expressed with following \*\* 7 is characterized by having the structure where the bridge was constructed in the bridge formation machine.

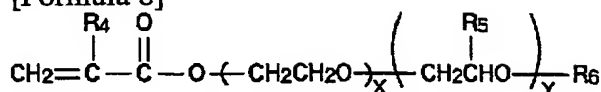


(R<sub>1</sub>, R<sub>2</sub>, and R<sub>3</sub> express a hydrogen atom or an alkyl group among a formula, and x and y are the integers of x >= 1 and y >= 0 or x >= 0, and y >= 1.)

[Claim 23] The compound by which bridge formation was carried out [ aforementioned ] is an electrolyte according to claim 22 with which the monoester compound expressed with \*\* 8 of further the following is characterized by having the structure where the bridge was constructed in the bridge .../tran\_web\_cgi\_ejje?u=http%3A%2F%2Fwww6.ipdl.jpo.go.jp%2FTokujitu%2Ftjitemcnt.ipdl 6/4/2003

formation machine.

[Formula 8]

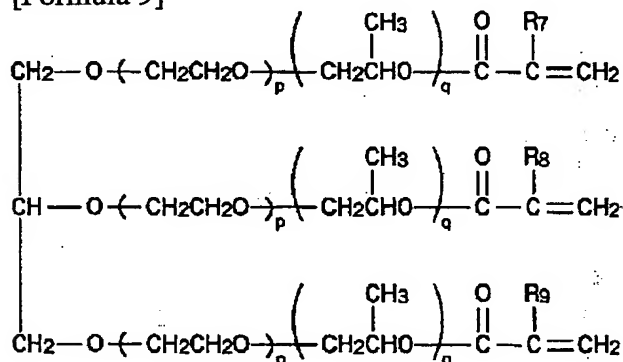


(R4, R5, and R6 express a hydrogen atom or an alkyl group among a formula, and X and Y are the integers of  $X \geq 1$  and  $Y \geq 0$  or  $X = 0$ , and  $Y \geq 1$ .)

[Claim 24] The compound by which bridge formation was carried out [aforementioned] is an electrolyte according to claim 23 characterized by having more greatly than 0 the structure where the bridge was constructed over the diester compound in the bridge formation machine, and the structure where the bridge was constructed over the monoester compound in the bridge formation machine, within the limits of 5.0 or less in the weight ratio (a monoester compound / diester compound) of a monoester compound to a diester compound.

[Claim 25] The compound by which bridge formation was carried out [aforementioned] is an electrolyte according to claim 21 with which the triester compound expressed with following \*\* 9 is characterized by having the structure where the bridge was constructed in the bridge formation machine.

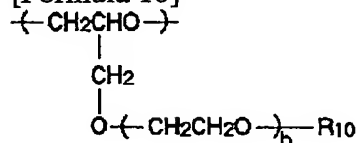
[Formula 9]



(R7, R8, and R9 express a hydrogen atom or an alkyl group among a formula, and p and q are the integers of  $p \geq 1$  and  $q \geq 0$  or  $p = 0$ , and  $q \geq 1$ .)

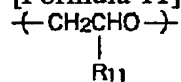
[Claim 26] The compound by which bridge formation was carried out [aforementioned] is an electrolyte according to claim 17 with which a copolymer including the composition unit expressed with the composition unit expressed with following \*\* 10 and following \*\* 11 is characterized by having the structure where the bridge was constructed in the bridge formation machine.

[Formula 10]



(R10 expresses at least one sort in an alkyl group, an alkenyl machine, a cycloalkyl machine, an aryl group, an aralkyl machine, and a tetrahydropyranyl group among a formula, and h is the integer of  $1 \leq h \leq 12$ .)

[Formula 11]

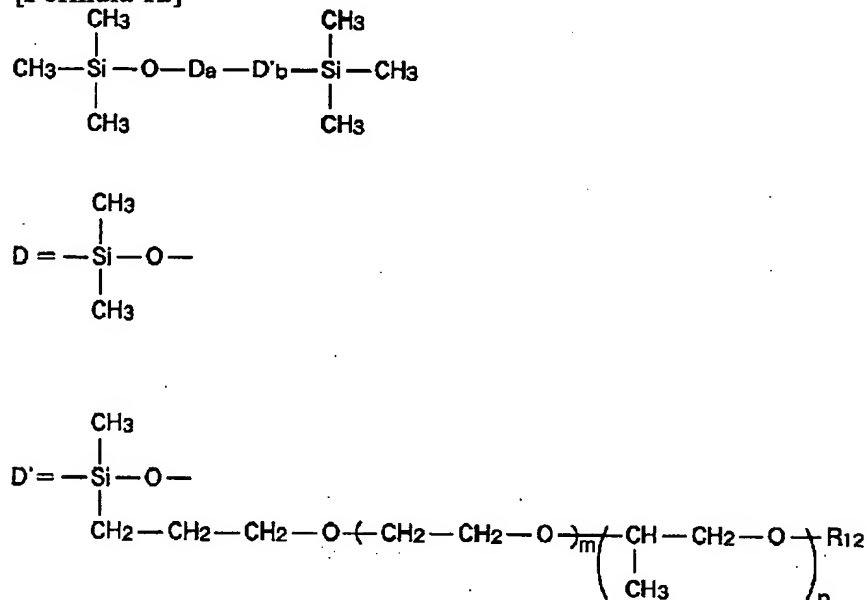


(the inside of the functional group in which R11 has an active hydrogen machine and an unsaturation double bond among a formula -- at least -- on the other hand -- or the inside of the functional group which has an active hydrogen machine and an unsaturation double bond -- on the other hand, at least one sort in a hydrogen atom, an alkyl group, an alkenyl machine, a cycloalkyl machine, and an aryl group is expressed to a list at least)

[Claim 27] The rate of the aforementioned electrolyte salt to the compound by which bridge formation was carried out [ aforementioned ] is an electrolyte according to claim 17 characterized by being the rate (the number of mols of the ether linkage unit in the number of mols / compound with which the bridge was constructed of an electrolyte salt) of the number of mols of an electrolyte salt to the number of mols of the ether linkage unit in the compound over which the bridge was constructed, and being within the limits of five or less [ 0.0001 or more ].

[Claim 28] The aforementioned high molecular compound is an electrolyte according to claim 17 characterized by including the siloxane derivative expressed with following \*\* 12.

[Formula 12]



(a expresses the integer of 0 to 100 among a formula, b expresses the integer of 1 to 100, m expresses the integer of 0 to 100, n expresses the integer of 0 to 100, and R<sub>12</sub> expresses a hydrogen atom or an alkyl group.) In addition, at the time of b > 1, even if b D' is the same, they may differ. Moreover, the hydrogen atom contained in D' and R<sub>12</sub> may be replaced by the halogen atom.

[Claim 29] The rate of the aforementioned electrolyte salt to the aforementioned siloxane derivative is an electrolyte according to claim 28 characterized by being the rate (the number of mols of the ether linkage unit in the number of mols / siloxane derivative of an electrolyte salt) of the number of mols of an electrolyte salt to the number of mols of the ether linkage unit in a siloxane derivative, and being within the limits of five or less [ 0.0001 or more ].

[Claim 30] The electrolyte according to claim 28 characterized by including the compound by which bridge formation was carried out [ aforementioned ] to the 100 weight sections of the aforementioned siloxane derivative by within the limits below the 10000 weight sections more than 10 weight sections.

[Claim 31] The average molecular weight of the aforementioned siloxane derivative is an electrolyte according to claim 28 characterized by being 100000 or less.

[Claim 32] The aforementioned electrolyte salt is an electrolyte according to claim 17 characterized by being lithium salt.

[Claim 33] The manufacture method of the electrolyte characterized by mixing the compound which has the functional group in which ether linkage and bridge formation are possible, and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt, and making the compound which can construct a bridge construct a bridge.

[Claim 34] The manufacture method of the electrolyte according to claim 33 characterized by making the compound which can construct a bridge irradiating ultraviolet rays, an electron ray, X-rays, gamma rays, microwave, or a RF or by heating construct a bridge.

[Claim 35] The manufacture method of the electrolyte characterized by adding an electrolyte salt after mixing the compound which has the functional group in which ether linkage and bridge formation are possible, and for which a bridge can be constructed, and a high molecular compound and making the

compound which can construct a bridge construct a bridge.

[Claim 36] The manufacture method of the electrolyte according to claim 35 characterized by making the compound which can construct a bridge irradiating ultraviolet rays, an electron ray, X-rays, gamma rays, microwave, or a RF or by heating construct a bridge.

[Claim 37] It is the cell characterized by including the compound with which it is the cell equipped with the electrolyte with the positive electrode and the negative electrode, and the bridge was constructed over the compound with which the aforementioned electrolyte has ether linkage and a bridge formation machine in the bridge formation machine, a high molecular compound, and an electrolyte salt.

[Claim 38] The compound by which bridge formation was carried out [ aforementioned ] is a cell according to claim 37 by which one sort which has at least one bridge formation machine, or two sorts or more of compounds are characterized by having the structure where the bridge was constructed in the bridge formation machine.

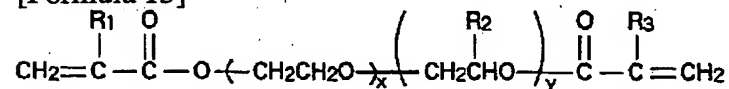
[Claim 39] The compound by which bridge formation was carried out [ aforementioned ] is an electrolyte according to claim 37 characterized by having a free end side chain including ether linkage.

[Claim 40] The compound by which bridge formation was carried out [ aforementioned ] is a cell according to claim 37 by which an ester compound is characterized by having the structure where the bridge was constructed in the bridge formation machine.

[Claim 41] The compound by which bridge formation was carried out [ aforementioned ] is a cell according to claim 40 characterized by having the structure where the bridge was constructed over at least one sort in a monoester compound, a diester compound, and a triester compound in the bridge formation machine.

[Claim 42] The compound by which bridge formation was carried out [ aforementioned ] is a cell according to claim 41 by which the diester compound expressed with following \*\* 13 is characterized by having the structure where the bridge was constructed in the bridge formation machine.

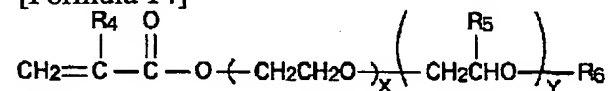
[Formula 13]



(R1, R2, and R3 express a hydrogen atom or an alkyl group among a formula, and x and y are the integers of  $x \geq 1$  and  $y \geq 0$  or  $x \geq 0$ , and  $y \geq 1$ .)

[Claim 43] The compound by which bridge formation was carried out [ aforementioned ] is a cell according to claim 42 by which the monoester compound expressed with \*\* 14 of further the following is characterized by having the structure where the bridge was constructed in the bridge formation machine.

[Formula 14]

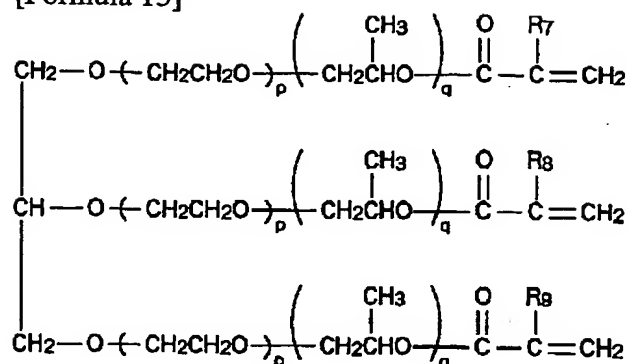


(R4, R5, and R6 express a hydrogen atom or an alkyl group among a formula, and X and Y are the integers of  $X \geq 1$  and  $Y \geq 0$  or  $X \geq 0$ , and  $Y \geq 1$ .)

[Claim 44] The compound by which bridge formation was carried out [ aforementioned ] is a cell according to claim 43 characterized by having more greatly than 0 the structure where the bridge was constructed over the diester compound in the bridge formation machine, and the structure where the bridge was constructed over the monoester compound in the bridge formation machine, within the limits of 5.0 or less in the weight ratio (a monoester compound / diester compound) of a monoester compound to a diester compound.

[Claim 45] The compound by which bridge formation was carried out [ aforementioned ] is a cell according to claim 41 by which the triester compound expressed with following \*\* 15 is characterized by having the structure where the bridge was constructed in the bridge formation machine.

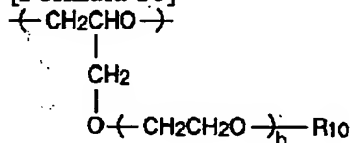
[Formula 15]



(R7, R8, and R9 express a hydrogen atom or an alkyl group among a formula, and p and q are the integers of  $p \geq 1$  and  $q \geq 0$  or  $p \geq 0$ , and  $q \geq 1$ .)

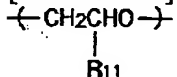
[Claim 46] The compound by which bridge formation was carried out [aforementioned] is a cell according to claim 37 by which a copolymer including the composition unit expressed with the composition unit expressed with following \*\* 16 and following \*\* 17 is characterized by having the structure where the bridge was constructed in the bridge formation machine.

[Formula 16]



(R10 expresses at least one sort in an alkyl group, an alkenyl machine, a cycloalkyl machine, an aryl group, an aralkyl machine, and a tetrahydropyranyl group among a formula, and h is the integer of  $1 \leq h \leq 12$ .)

[Formula 17]

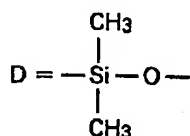
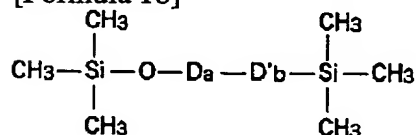


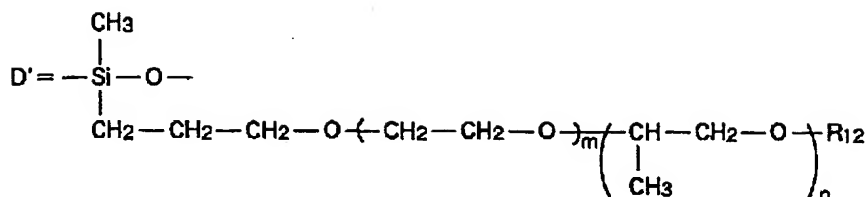
(the inside of the functional group in which R11 has an active hydrogen machine and an unsaturation double bond among a formula -- at least -- on the other hand -- or the inside of the functional group which has an active hydrogen machine and an unsaturation double bond -- on the other hand, at least one sort in a hydrogen atom, an alkyl group, an alkenyl machine, a cycloalkyl machine, and an aryl group is expressed to a list at least)

[Claim 47] The rate of the aforementioned electrolyte salt to the compound by which bridge formation was carried out [aforementioned] is a cell according to claim 37 characterized by being the rate (the number of mols of the ether linkage unit in the number of mols / compound with which the bridge was constructed of an electrolyte salt) of the number of mols of an electrolyte salt to the number of mols of the ether linkage unit in the compound over which the bridge was constructed, and being within the limits of five or less [0.0001 or more].

[Claim 48] The aforementioned high molecular compound is a cell according to claim 37 characterized by including the siloxane derivative expressed with following \*\* 18.

[Formula 18]





(a expresses the integer of 0 to 100 among a formula, b expresses the integer of 1 to 100, m expresses the integer of 0 to 100, n expresses the integer of 0 to 100, and R<sub>12</sub> expresses a hydrogen atom or an alkyl group.) In addition, at the time of b > 1, even if b D' is the same, they may differ. Moreover, the hydrogen atom contained in D' and R<sub>12</sub> may be replaced by the halogen atom.

[Claim 49] The rate of the aforementioned electrolyte salt to the aforementioned siloxane derivative is a cell according to claim 48 characterized by being the rate (the number of mols of the ether linkage unit in the number of mols / siloxane derivative of an electrolyte salt) of the number of mols of an electrolyte salt to the number of mols of the ether linkage unit in a siloxane derivative, and being within the limits of five or less [ 0.0001 or more ].

[Claim 50] The cell according to claim 48 characterized by including the compound by which bridge formation was carried out [ aforementioned ] to the 100 weight sections of the aforementioned siloxane derivative by within the limits below the 10000 weight sections more than 10 weight sections.

[Claim 51] The average molecular weight of the aforementioned siloxane derivative is a cell according to claim 48 characterized by being 100000 or less.

[Claim 52] The aforementioned electrolyte salt is a cell according to claim 37 characterized by being lithium salt.

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[Translation done.]



## \* NOTICES \*

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1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

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**DETAILED DESCRIPTION**


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[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] this invention relates to the cell which used an electrolyte, its manufacture method, and it for the constituent row for electrolytes containing a high molecular compound and an electrolyte salt.

[0002]

[Description of the Prior Art] Recent years come, portable electrical-and-electric-equipment products, such as a camcorder/movie (video tape recorder), a cellular phone, or a laptop computer, are spreading quickly, and the further high performance-ization of an electrochemistry device is needed as a device of such electronic equipment.

[0003] Conventionally, the liquefied electrolyte made to dissolve an electrolyte salt in water or an inflammable organic solvent has been used as matter with which electrochemistry devices, such as a rechargeable battery, manage ionic conduction. However, a liquefied electrolyte has problems, such as a liquid spill, and, for this reason, needed to secure confidentiality using the metal container. Therefore, generally, such an electrochemistry device had the heavy weight, and the flexibility of a configuration also had complicatedness low [ a device ] in connection with the sealing process. Then, the research using the so-called solid electrolyte which consists of an ion conductivity solid-state is made actively. While there are no worries about a liquid spill in a solid electrolyte and being able to carry out simple [ of the sealing process for it ], there is an advantage, like the flexibility of shape selectivity is high by the film moldability which can carry out [ lightweight ]-izing of the device and which was further excellent in the polymer.

[0004] Generally this solid electrolyte consists of a matrix macromolecule and a possible electrolyte salt of ionic dissociation. Among these, the matrix macromolecule holds ionic dissociation ability and has the function of both sides, such as a duty which solidifies this ion conductivity solid-state, and a duty as a solvent to an electrolyte salt. About such a solid electrolyte, the report that the ionic conductivity of about  $1 \times 10^{-7}$  S/cm was obtained by the system which dissolved the lithium perchlorate in the polyethylene oxide by Armand and others of the Grenoble university (France) is carried out in 1978, and examination has also been performed still more actively since then about the various polymeric materials focusing on the polymer which has polyether combination now.

[0005]

[Problem(s) to be Solved by the Invention] The solid electrolyte using a straight chain-like polyether which is represented by this polyethylene oxide as a matrix shows ionic conduction, when the ion which is dissolving moves by the local segmental motion of a macromolecule chain in the inside of the amorphous phase more than the glass transition temperature of a matrix polymer.

[0006] However, the ion which is dissolving into the matrix of the shape of a straight chain like the polyethylene oxide which is the polymer of semicrystallinity, especially the cation were strongly configured according to the interaction with a macromolecule chain, this became a point constructing [ false ] a bridge, and they caused partial crystallization, and had the problem that a segmental motion will fall. Therefore, in order to realize higher ionic conductivity under a room temperature, to perform a molecular design which the dissociating power of an electrolyte salt is high, and the amorphism (amorphous) field where ion tends to move within a matrix exists mostly, and keeps the glass transition point of a polymer low is desired.

[0007] In addition, the attempt which raises ionic conductivity by introducing branching structure into a polyethylene-oxide skeleton as this kind of a molecular design, for example is performed (21 the Watanabe justice, Netsu Sokutei 24 (1) pp12- 1996). However, this kind of polymer has the problem that the synthetic method is complicated.

[0008] Moreover, otherwise, the three-dimensions network structure is introduced into a matrix macromolecule, the attempt which makes crystallization of a polymer check is made, and the method of carrying out the polymerization of the acrylic or the methacrylic system monomer which has a polyoxyalkylene component as an example of this kind of molecular design is reported (JP,5-25353,A). However, there is a problem that ionic conductivity for a low sake with the sufficient melting nature to the monomer of an alkali-metal salt is not obtained. Therefore, the new solid electrolyte replaced with these is called for.

[0009] this invention was made in view of this trouble, and the purpose is in offering the cell which used an electrolyte, its manufacture method, and it for the constituent row for electrolytes which can show high ion conductivity.

[0010]

[Means for Solving the Problem] The constituent for electrolytes by this invention contains the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt.

[0011] The compound with which the electrolyte by this invention has ether linkage and a bridge formation machine contains the compound over which the bridge was constructed in the bridge formation machine, a high molecular compound, and an electrolyte salt.

[0012] The manufacture method of the electrolyte by this invention mixes the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt, and makes the compound which can construct a bridge construct a bridge.

[0013] After the manufacture method of other electrolytes by this invention mixes the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, and a high molecular compound and makes the compound which can construct a bridge construct a bridge, it adds an electrolyte salt.

[0014] The cell by this invention is equipped with an electrolyte with a positive electrode and a negative electrode, and an electrolyte contains the compound with which the bridge was constructed over the compound which has ether linkage and a bridge formation machine in the bridge formation machine, a high molecular compound, and an electrolyte salt.

[0015] In the constituent for electrolytes by this invention, since the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, and the high molecular compound are included, the electrolyte in which high ion conductivity is shown is obtained.

[0016] In the electrolyte by this invention, the electrolyte salt dissociated with the high molecular compound between the compounds with which the bridge was constructed over the compound which has ether linkage and a bridge formation machine in the bridge formation machine exists. Therefore, high ion conductivity is shown.

[0017] By the manufacture method of the electrolyte by this invention, after the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt are mixed, a bridge is constructed over the compound for which a bridge can be constructed.

[0018] By the manufacture method of the electrolyte by this invention, after the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, and a high molecular compound are mixed and the polymerization of the compound which can construct a bridge is carried out, an electrolyte salt is added.

[0019] By the cell by this invention, when the ion produced by the maceration of an electrolyte salt moves between a negative electrode and a positive electrode in the inside of an electrolyte, it discharges. Here, since it has the electrolyte of this invention, it has high ion conductivity and the outstanding cell performance is shown.

[0020]

[Embodiments of the Invention] Hereafter, the gestalt of operation of this invention is explained in detail.

[0021] The electrolyte concerning the gestalt of 1 operation of this invention contains the compound which has the three-dimensions network structure by which the bridge was constructed over the compound which has ether linkage and a bridge formation machine in the bridge formation machine, the high molecular compound, and the electrolyte salt. Thus, the compound which has the three-dimensions network structure and over which the bridge was constructed is included because membranes cannot be formed since a fluidity is shown as molecular weight becomes low, and it is hard to obtain sufficient film intensity only with a high molecular compound also in a high molecular weight object. That is, the compound which has this three-dimensions network structure and over which the bridge was constructed is for holding a high molecular compound and an electrolyte salt between the three-dimensions network structure, and collateralizing membrane formation nature and strong film intensity.

[0022] As this compound over which the bridge was constructed, what has the structure where the bridge was constructed over either [ at least ] the straight chain-like compound or the compound of the pectinate in the bridge formation machine is mentioned, and you may have the side chain, for example. In addition, this compound over which the bridge was constructed is not restricted to that by which the bridge was constructed over the open chain compound that what is necessary is just to have the structure where the bridge was constructed over the compound which contains a chain-like atomic arrangement in part at least for example, but may have the structure where the bridge was constructed over the compound which has an annular atomic arrangement in part. That is, this compound over which the bridge was constructed may have the annular atomic arrangement in the principal chain or the side chain.

[0023] Moreover, a bridge may be constructed over the compound with which the compound with which this compound over which the bridge was constructed has at least one bridge formation machine has a bridge formation machine in what position that what is necessary is just to construct a bridge. For example, although the bridge was constructed over the straight chain-like compound, the bridge may be constructed over the compound which has a bridge formation machine to ends, and may be constructed over the compound which has a bridge formation machine at one end at the case. Although the bridge was constructed over the compound of the pectinate, the bridge may be constructed over the compound which has a bridge formation machine at at least one end at the case. Namely, this compound over which the bridge was constructed should just form the three-dimensions network structure which can hold a high molecular compound and an electrolyte salt.

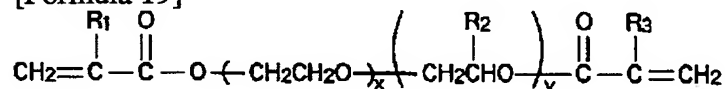
[0024] However, in order to realize higher ionic conductivity, it is desirable to have the structure where the bridge was constructed over the compound which has at least one free end which a bridge formation machine is not arranged and includes ether linkage. It is because it will have the free end side chain which branching structure is introduced into the three-dimensions network structure by this, and includes ether linkage. For example, when constituted by the compound with which the bridge was constructed over the straight chain-like compound, it is desirable to have the structure where the bridge was constructed over the compound which has a bridge formation machine at one end in addition to the structure where the bridge was constructed over the compound which has a bridge formation machine to ends. Moreover, when constituted by the compound with which the bridge was constructed over the compound of the pectinate, it is desirable to have the structure where the bridge was constructed over the compound of the pectinate which has at least one free end on which a bridge formation machine is not arranged. Furthermore, when constituted by the compound with which the bridge was constructed over the straight chain-like compound and the compound of the pectinate, it is desirable to have the structure where the bridge was constructed over the compound of the shape of a straight chain which has a bridge formation machine at one end, or the structure where the bridge was constructed over the compound of the pectinate which has at least one free end which is not matched with a bridge formation machine.

[0025] This compound over which the bridge was constructed may have the structure of cross linkage by combination like a throat, such as ether linkage, ester combination, or a urethane bond, again. For

example, the compound which has an allyl group, a vinyl group, an acrylate machine, a methacrylate machine, the functional group that has cycloolefin structure, or an active hydrogen machine may have the structure of cross linkage over which the bridge was constructed in them. Moreover, you may have the structure of cross linkage from which plurality differs in 1 compound.

[0026] Specifically as such a compound over which the bridge was constructed, what has the structure where what has the structure of cross linkage by which the bridge was constructed over the ester compound in the bridge formation machine was mentioned, and the bridge was constructed over at least one sort in a monoester compound, a diester compound, and a triester compound especially is desirable. For example, what has the structure where the bridge was constructed over the diester compound including the oxy-alkylene structure expressed with the general formula shown in \*\* 19 as a compound which has the structure where the bridge was constructed over the diester compound in the bridge formation machine is mentioned.

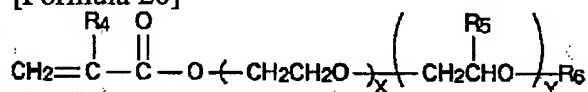
[Formula 19]



The inside of a formula, R1, and R2 And R3 A hydrogen atom or the alkyl group of carbon numbers 1-6 is expressed, and even if each is the same, it may differ. Moreover, x and y are the integers of  $x \geq 1$  and  $y \geq 0$  or  $x = 0$ , and  $y \geq 1$ .

[0027] In this case, it is more desirable to have the structure where the bridge was constructed over the monoester compound with which the diester compound expressed with the general formula shown in \*\* 19, for example includes the oxy-alkylene structure which is expressed with the general formula shown in \*\* 20 in addition to the structure where the bridge was constructed in the bridge formation machine in the bridge formation machine. It is because a free end side chain including ether linkage is introduced and higher ionic conductivity can be obtained, as mentioned above.

[Formula 20]

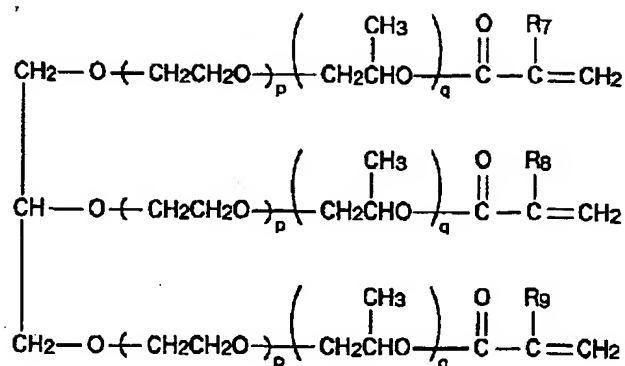


The inside of a formula, R4, and R5 And R6 A hydrogen atom or the alkyl group of carbon numbers 1-6 is expressed, and even if each is the same, it may differ. Moreover, X and Y are the integers of  $X \geq 1$  and  $Y \geq 0$  or  $X = 0$ , and  $Y \geq 1$ .

[0028] In this case, as for the structure where the bridge was constructed over the diester compound in the bridge formation machine, and the structure where the bridge was constructed over the monoester compound in the bridge formation machine, in the weight ratio (a monoester compound / diester compound) of a monoester compound to a diester compound, it is desirable that it is within the limits of 5.0 or less more greatly than 0. It is because the three-dimensions network structure cannot be constituted if there is little structure where the bridge was constructed over the diester compound, but the free end side chain which includes ether linkage when there is little structure where the bridge was constructed over the monoester compound decreases and ionic conductivity cannot be made high.

[0029] Moreover, what has the structure where the bridge was constructed over the triester compound including the oxy-alkylene structure expressed with the general formula shown in \*\* 21 as a compound which has the structure where the bridge was constructed over the triester compound, for example in the bridge formation machine is mentioned.

[Formula 21]



The inside of a formula, R7, and R8 And R9 A hydrogen atom or the alkyl group of carbon numbers 1-6 is expressed, and even if each is the same, it may differ. Moreover, p and q are the integers of  $p \geq 1$  and  $q \geq 0$  or  $p \geq 0$ , and  $q \geq 1$ .

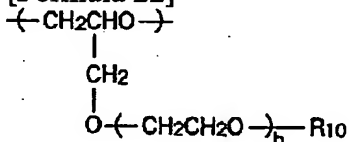
[0030] Although especially the composition ratio of the oxyethylene unit of the triester compound shown in this \*\* 21 and an oxypropylene unit is not limited, it is desirable that it is within the limits of  $0.1 \leq q/p \leq 4$ . It is because it will become weak if many [ if there are too few oxyethylene units, a mechanical strength will fall, and / too ]. Moreover, although any of the shape of the letter of a block or random are sufficient as the joint format of an oxyethylene unit and an oxypropylene unit, the shape of random is more desirable.

[0031] It is more desirable to have the structure where the bridge was constructed over the monoester compound expressed with the general formula shown in \*\* 20 in the bridge formation machine in addition to the structure where the bridge was constructed over the triester compound expressed with the general formula shown in \*\* 21 in the bridge formation machine, as mentioned above also in this case.

[0032] moreover -- as the compound over which the bridge was constructed -- these -- others -- for example, what has the structure where the bridge was constructed over the copolymer including the composition unit shown in the composition unit shown in \*\* 22 and \*\* 23 in the bridge formation machine is mentioned Since according to this compound the free end side chain which includes ether linkage by the composition unit shown in \*\* 22 is introduced and high ionic conductivity can be obtained, it is desirable.

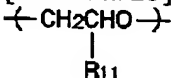
[0033]

[Formula 22]



R10 expresses at least one sort in the alkyl group of carbon numbers 1-12, the alkenyl machine of carbon numbers 2-18, the cycloalkyl machine of carbon numbers 3-8, the aryl group of carbon numbers 6-18, the aralkyl machine of carbon numbers 7-12, and a tetrahydropyranyl group among a formula, and h is the integer of  $1 \leq h \leq 12$ . In addition, R10 may express the same content in all composition units, and may express the content which changes with composition units.

[Formula 23]



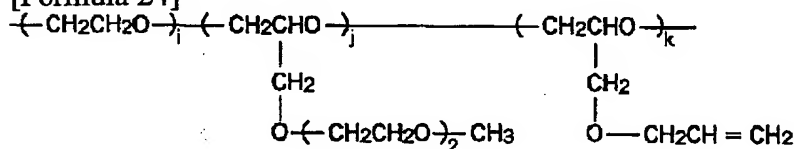
R11 expresses among a formula at least one sort in at least one side of the functional groups which have at least one side of the functional groups which have an active hydrogen machine and an unsaturation double bond or an active hydrogen machine, and an unsaturation double bond, a hydrogen atom, an alkyl group, an alkenyl machine, a cycloalkyl machine, and an aryl group. In addition, R11 may express the same content in all composition units, and may express the content which changes with composition units.

[0034] Any of the shape of the letter of a block or random are sufficient as the joint format with the

composition unit shown in the composition unit which especially the composition ratio with the composition unit shown in the composition unit shown in \*\* 22 in this copolymer and \*\* 23 is not limited, and was shown in \*\* 22, and \*\* 23. Incidentally, in this copolymer, the functional group which can construct a bridge is a functional group which has an active hydrogen machine and an unsaturation double bond. As a functional group which has an unsaturation double bond, the functional group which has an allyl group, a vinyl group, an acrylate machine, a methacrylate machine, or cycloolefin structure is mentioned.

[0035] That by which the bridge was constructed over the copolymer specifically expressed with the general formula shown in \*\* 24 as a compound which has such structure, and over which the bridge was constructed in the bridge formation machine is desirable. In \*\* 22, as for the copolymer shown in \*\* 24, k includes [R10 / R11] the composition unit which consists of an allyl group in the composition unit which consists of a methyl group, the composition unit which R11 becomes from a hydrogen atom in \*\* 23, and \*\* 23 by two.

[Formula 24]



i, j, and k are one or more integers among a formula.

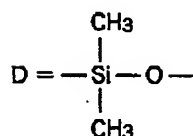
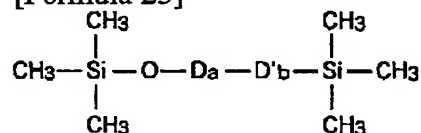
[0036] In addition, you may mix and use two or more compounds with which such structures differ for this compound over which the bridge was constructed. For example, you may mix and use the compound which has the structure where the bridge was constructed over the copolymer including the compound which has the structure where the bridge was constructed over the ester compound shown in \*\* 19, and the composition unit shown in the composition unit shown in \*\* 22, and \*\* 23.

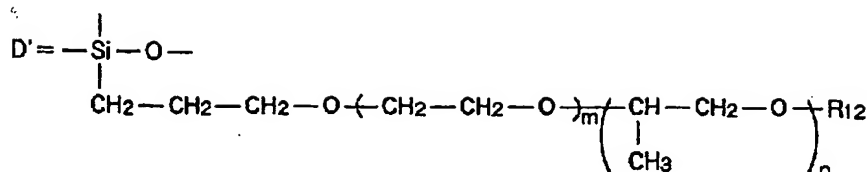
[0037] A high molecular compound is for dissolving an electrolyte salt. It is n (SiOCH<sub>3</sub> R) which has the chain combination of silicon (Si) and oxygen (O) in a basic skeleton and by which the side chain R was added to silicon as this high molecular compound, for example. The shape type siloxane derivative of a chain expressed is mentioned. In this case, a substituent or a side chain radical R is a monovalent organic machine, and serves as the structure where an electrolyte salt can be dissolved, by choosing a side chain radical R moderately. Therefore, as for the average molecular weight of this siloxane derivative, it is desirable that it is 100000 or less.

[0038] Moreover, since high compatibility with the compound with which the bridge was constructed over the compound which has ether linkage in the bridge formation machine is needed while dissolving an electrolyte salt effectively, as for this siloxane derivative, it is desirable to include ether linkage in a substituent or a side chain radical R. As such a side chain radical R, there are alkoxy groups, such as an ethoxy basis, a propoxy group, a butoxy machine, and a methoxy machine. Moreover, the hydrogen in a side chain radical R may be replaced by halogens, such as boron or a fluorine.

[0039] What is specifically expressed with the general formula shown in \*\* 25 as such a siloxane derivative is desirable.

[Formula 25]





a expresses the integer of 0 to 100 among a formula, b expresses the integer of 1 to 100, m expresses the integer of 0 to 100, n expresses the integer of 0 to 100, and R12 expresses a hydrogen atom or an alkyl group. In addition, at the time of  $b > 1$ , even if b D' is the same, they may differ. Moreover, the hydrogen atom contained in D' and R12 may be replaced by the halogen atom.

[0040] An electrolyte salt is for giving ion conductivity by maceration, and is expressed with the general formula shown in \*\* 26.

[Formula 26] A expresses a cation among AB formula and B expresses an anion.

[0041] A light metal salt etc. is mentioned as this electrolyte salt, and, specifically, it is used according to the purpose from alkaline-earth-metals salts, such as alkali-metal salts, such as a lithium (Li) salt, a sodium (Na) salt, or a potassium (K) salt, a calcium (calcium) salt, or a magnesium (Mg) salt, etc. In addition As lithium salt \*\*  $\text{LiClO}_4$ ,  $\text{LiBF}_4$ ,  $\text{LiPF}_6$ ,  $\text{LiAsF}_6$ ,  $\text{LiAlCl}_4$ ,  $\text{LiSbF}_6$ ,  $\text{LiCF}_3\text{SO}_3$ ,  $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ ,  $\text{LiC}_4\text{F}_9\text{SO}_3$ ,  $\text{LiCF}_3\text{CO}_2$ , and  $\text{LiN}(\text{CF}_3\text{CO}_2)_2$  It is. etc. -- as sodium salt --  $\text{NaClO}_4$ ,  $\text{NaBF}_4$ ,  $\text{NaSCN}$ , etc. -- it is -- as potassium salt --  $\text{KBF}_4$  etc. -- it is The need is accepted, a gap, one sort, or two sorts or more are mixed, and these are used.

[0042] In addition, when a siloxane derivative is used as a high molecular compound, as for the rate of the compound and high molecular compound over which the bridge was constructed, it is desirable to make into within the limits below the 10000 weight sections the compound over which the bridge was constructed to the 100 weight sections of a siloxane derivative more than 10 weight sections. It is because un-arranging -- a film becomes easily or hard -- will arise if film intensity is not enough if there are few compounds over which the bridge was constructed than 10 weight sections, and the 10000 weight sections are exceeded, although based also on the molecular weight of a siloxane derivative, or an electrolytic use.

[0043] Moreover, the rate of the electrolyte salt to the compound over which the bridge was constructed is a rate (the number of mols of the ether linkage unit in the number of mols / compound with which the bridge was constructed of an electrolyte salt) of the number of mols of an electrolyte salt to the number of mols of the ether linkage unit in the compound over which the bridge was constructed, and it is desirable that it is within the limits of five or less [ 0.0001 or more ]. As for the rate of the electrolyte salt to a siloxane derivative, it is desirable similarly that it is [ of the number of mols of the electrolyte salt to the number of mols of the ether linkage unit in a siloxane derivative ] within the limits of five or less [ 0.0001 or more ] comparatively (the number of mols of the ether linkage unit in the number of mols / siloxane derivative of an electrolyte salt). It is because high ionic conductivity can be obtained within the limits of this.

[0044] The number of mols of the ether linkage unit incidentally included in one mol of diester compounds shown in \*\* 19 is mol  $(1+x+y)$ , and the number of mols of the ether linkage unit included in one mol of copolymers shown in \*\* 24 is  $\{i+(1+1+2)x_j+(1+1)x_k\}$  mol.

[0045] The electrolyte which has such composition can be manufactured as follows using the following constituents for electrolytes.

[0046] First, the constituent for electrolytes containing the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt is prepared. Among these, a high molecular compound and an electrolyte salt are as having mentioned above. For example, a siloxane derivative is prepared as a high molecular compound, and at least one sort of lithium salt is prepared as an electrolyte salt.

[0047] The compound which can construct a bridge turns into a compound which has the three-dimensions network structure mentioned above according to bridge formation and over which the bridge was constructed, the compounds or those mixture of the pectinate are sufficient also as a straight chain-like compound, and they may have the side chain. Moreover, as long as it has the chain-like atomic arrangement in part at least, the compound which has an atomic arrangement also



with an open chain compound annular to a part of principal chain or side chain is sufficient. Furthermore, if it has at least one functional group which can construct a bridge, any number of numbers are good and the position is also good anywhere. For example, in the case of the straight chain-like compound, you may have to ends the functional group which can construct a bridge, and may have only at one end. You may be the many organic functions which may have the functional group which can construct a bridge over at least one end in the case of the compound of the pectinate, and it has three or more.

[0048] Moreover, the compound which can construct a bridge may have what functional group as a functional group which can construct a bridge. Specifically, the compound which has an ester compound or an allyl group is mentioned. For example, as long as it is an ester compound, you may have a monoester compound, a diester compound, a triester compound, or ester combination four or more. Furthermore, the compound in which this bridge formation is possible may contain two or more sorts of things from which the number of the functional groups which can construct a bridge differs, and may contain two or more sorts of things from which the kind of functional group which can construct a bridge differs.

[0049] The triester compound expressed with the general formula shown in the monoester compound and \*\* 21 which are expressed with the diester compound expressed with the general formula shown in \*\* 19 as a compound in which such bridge formation is possible, for example, and the general formula shown in \*\* 20 is mentioned. In addition, it is more desirable to include the monoester compound shown in \*\* 20 in addition to this compound, when using the diester compound shown in \*\* 19. As mentioned above, when this constructs a bridge, it is for forming a free end side chain including ether linkage. In this case, as for the weight ratio (a monoester compound / diester compound) of a monoester compound to a diester compound, it is desirable that it is within the limits of 5.0 or less more greatly than 0. Moreover, it is more desirable to include the monoester compound shown in \*\* 20 in addition to this compound, when using the triester compound shown in \*\* 21.

[0050] A copolymer including the composition unit shown in the composition unit shown in \*\* 22 and \*\* 23 as a compound which can construct a bridge over others, for example etc. is mentioned. Specifically, the copolymer shown in \*\* 24 is desirable.

[0051] In addition, it is desirable to make into within the limits below the 10000 weight sections the compound which can construct a bridge more than 10 weight sections to the 100 weight sections of a siloxane derivative as the rate of the compound and high molecular compound which can construct a bridge was mentioned above, when a siloxane derivative was used as a high molecular compound. Moreover, the rate of the electrolyte salt to the compound which can construct a bridge as mentioned above The number of mols of the electrolyte salt to the number of mols of the ether linkage unit in the compound which can construct a bridge comparatively (the number of mols of the ether linkage unit in the compound in which electrolytic number of mols / bridge formation are possible) The rate of the aforementioned electrolyte salt [ as opposed to / it is desirable that it is within the limits of five or less / 0.0001 or more / , and / a siloxane derivative ] It is desirable that it is [ of the number of mols of the electrolyte salt to the number of mols of the ether linkage unit in a siloxane derivative ] within the limits of five or less [ 0.0001 or more ] comparatively (the number of mols of the ether linkage unit in electrolytic number of mols / siloxane derivative).

[0052] Subsequently, the compound which has the functional group in which these ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt are mixed, and uniform mixture is produced. In addition, a polymerization initiator is also added if needed in that case. A polymerization initiator is needed except when irradiation of an electron ray performs a polymerization, and it has a photopolymerization initiator and a thermal polymerization initiator.

[0053] As a photopolymerization initiator, there is acetophenone, TORIKURORO acetophenone, 2-hydroxy-2-methylpropiohenone, 2-hydroxy-2-methyl iso propiophenone, 1-hydroxy cyclohexyl ketone, BENZO iso ether, 2, and 2-diethoxy acetophenone or a benzyl dimethyl ketal, for example. Moreover, as a thermal polymerization initiator, there is initiators, such as elevated-temperature polymerization initiators, such as cumene hydroperoxide, t-butyl hydroperoxide, a JIKUMI loopier oxide, or G t-butyl peroxide, or a benzoyl peroxide, a lauroyl peroxide, a persulfate, or an



azobisisobutyronitril, or a redox initiator, for example. As a polymerization initiator, you may mix and use 1 of sorts of these, and two sorts or more.

[0054] In addition, a polymerization initiator is added in the range of the 0.1 - 1.0 weight section to the compound 100 weight section which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed. It is because an effect does not change even if a rate of polymerization will fall remarkably and will add exceeding the 1.0 weight sections, if fewer than the 0.1 weight section.

[0055] Then, ultraviolet rays, an electron ray, X-rays, gamma rays, microwave, or a RF is irradiated at this mixture, or this mixture is heated, and the polymerization of the compound which can construct a bridge is carried out. A bridge is constructed over the compound which can construct a bridge by this, and the electrolyte concerning the gestalt of this operation is obtained.

[0056] In addition, in case the compound which can construct a bridge, a high molecular compound, and an electrolyte salt are mixed, proper organic solvents, such as an acetonitrile, are used and you may make it form mixture. In this case, after performing dryness processing of leaving or heating in air and under reduced pressure after carrying out the polymerization of the compound which can construct a bridge, and making it remove an organic solvent, and obtaining the mixture, performing dryness processing and removing an organic solvent, you may be made to carry out the polymerization of the compound which can construct a bridge.

[0057] Moreover, this electrolyte can be manufactured even if it performs it as follows.

[0058] First, the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt are prepared like the previous manufacture method. Subsequently, it mixes with the compound which can construct a bridge, and a high molecular compound. A polymerization initiator is mixed if needed in that case. Then, the polymerization of the compound which can construct a bridge is carried out like the previous manufacture method, and the compound which can construct a bridge forms the mixture of the compound and high molecular compound over which the bridge was constructed in the bridge formation machine. Dissolve an electrolyte salt in the organic solvent after it, this mixture is made to permeate, and dryness removal of the organic solvent is carried out.

Thereby, the electrolyte concerning the gestalt of this operation is obtained.

[0059] In addition, although especially the conditions of polymerization reaction are not limited, polymerization reaction is considered from the simple nature of equipment, and a cost side, and UV irradiation or its heating polymerization is desirable. Moreover, when, producing the electrolyte used for the lithium cell using alkali-metal ion, a lithium ion battery, and a sodium cell for example, as for the solvent and polymerization initiator at the time of mixing the constituent for electrolytes (namely, the compound, high molecular compound, and alkali-metal salt which can construct a bridge), and these, it is desirable to fully perform dehydration processing, and it is desirable to also make atmosphere under manufacture into low humidity. It is because there are some which are decomposed depending on the kind of alkali-metal salt when this reacts with moisture. Moreover, it is because alkali metal and moisture react violently in a negative electrode when it uses as a cell.

[0060] Thus, the electrolyte manufactured is used for a cell as follows. Here, the example of the rechargeable battery which used the lithium is given and it explains below with reference to a drawing.

[0061] Drawing 1 expresses the cross-section structure of the rechargeable battery using the electrolyte concerning the gestalt of this operation. In addition, what was shown in drawing 1 is called so-called paper type. The laminating of this rechargeable battery is carried out through the electrolyte 13 which requires a negative electrode 11 and a positive electrode 12 for the gestalt of this operation. sheathing of the couple which the insulating packing 14 is arranged by these sides, and was arranged so that it might face across a negative electrode 11 and a positive electrode 12 in between -- it is sealed by pasting up with the periphery section of members 15 and 16

[0062] The negative electrode 11 contains occlusion and the metal which can be \*\*\*\*ed, the alloy, or the carbon material for for example, the lithium metal or the lithium ion. This carbon material is prepared in predetermined temperature and predetermined atmosphere, for example, carbon black, such as corks, such as pyrolytic carbons, petroleum coke, or pitch coke, artificial graphites, natural

graphites, and acetylene black, glassy carbons, the organic polymeric-materials baking object, or the carbon fiber is used. In addition, an organic polymeric-materials baking object calcinates organic polymeric materials at the suitable temperature of 500 degrees C or more in inert gas atmosphere or a vacuum.

[0063] a positive electrode 12 -- as a positive active material --  $\text{TiS}_2$ ,  $\text{MoS}_2$ , and  $\text{NbSe}_2$  Or  $\text{V}_2\text{O}_5$  etc. -- the metallic sulfide which does not contain a lithium, the oxide, the lithium compound sulfide containing a lithium, or the lithium multiple oxide is contained In order to make an energy density high especially, it is  $\text{Li}_x\text{MO}_2$ . It is desirable that the lithium multiple oxide made into a subject is included. In addition, one or more kinds of transition metals of M are desirable, and, specifically, its at least one sort in cobalt (Co), nickel (nickel), and manganese (Mn) is desirable. Moreover, x is usually the value of  $0.05 \leq x \leq 1.10$  within the limits. as the example of such a lithium multiple oxide --  $\text{LiCoO}_2$ ,  $\text{LiNiO}_2$ ,  $\text{Li}_x\text{Ni}_y\text{Co}_{1-y}\text{O}_2$ , or (however, the value of x and y changes with charge-and-discharge states of a cell, and are usually  $0 < x < 1$  and  $0.7 < y \leq 1$ .)  $\text{LiMn}_2\text{O}_4$  etc. -- it is mentioned

[0064] In addition, this lithium multiple oxide carries out trituration mixture according to composition of a request of the carbonate of a lithium, a nitrate, an oxide or a hydroxide, and the carbonate of transition metals, a nitrate, an oxide or a hydroxide, and is prepared by calcinating in oxygen atmosphere at the temperature within the limits of 600-1000 degrees C.

[0065] The electrolyte 13 contains lithium salt as an electrolyte salt here. Moreover, the electrolyte 13 serves also as the duty as separator in this rechargeable battery. That is, a negative electrode 11 and a positive electrode 12 are isolated, and a lithium ion is passed, preventing the short circuit of the current by contact of two poles. In addition, you may make it have the separator which is not illustrated between a positive electrode 12 and an electrolyte 13 or into an electrolyte 13 between a negative electrode 11 and an electrolyte 13 if needed. As separator, there is what consists of a nonwoven fabric made of synthetic resin, such as a polytetrafluoroethylene, polypropylene, or polyethylene, a thing which consists of a ceramic film, or a thing which consists of a porosity thin film film.

[0066] The rechargeable battery which has such composition acts as follows.

[0067] In this rechargeable battery, if it charges, a lithium will serve as ion, it will be desorbed from a positive electrode 12, and occlusion will be carried out to a negative electrode 11 through an electrolyte 13, for example. If it discharges, a lithium serves as ion and it is desorbed from a negative electrode 11, and through an electrolyte 13, a metaphor will return to a positive electrode 12 and occlusion will be carried out. Here, the compound which has ether linkage and a bridge formation machine contains the compound over which the bridge was constructed in the bridge formation machine, and the electrolyte 13 holds the high molecular compound and the lithium ion between the three-dimensions network structure. Therefore, while having membrane formation nature and strong film intensity, it also has high ion conductivity. Therefore, it has the outstanding cell performance.

[0068] Thus, since it was made for the compound which has ether linkage and a bridge formation machine to contain the compound and high molecular compound over which the bridge was constructed in the bridge formation machine according to the electrolyte concerning the gestalt of this operation, a high molecular compound and an electrolyte salt can be held between the three-dimensions network structure. Therefore, ion conductivity can be made high, holding high membrane formation nature and strong film intensity. Therefore, if an electrochemistry device is constituted using this electrolyte, the electrochemistry device which has a high performance easily can be obtained.

[0069] Moreover, if it is made to have the free end side chain in which the compound over which the bridge was constructed includes ether linkage, higher ionic conductivity can be obtained.

[0070] Furthermore, since it was made for the compound and high molecular compound which have the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed to be included according to the constituent for electrolytes concerning the gestalt of this operation, the polymerization of the compound in which this bridge formation is possible can be easily carried out by irradiation or heating of ultraviolet rays. Therefore, the electrolyte easily built over the gestalt of this operation can be obtained.

[0071] In addition, since it was made to carry out the polymerization of the compound for which a

bridge can be constructed after mixing the compound and high molecular compound which have the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed according to the manufacture method of the electrolyte concerning the gestalt of this operation, the electrolyte easily built over the gestalt of this operation can be obtained.

[0072] Furthermore, since the electrolyte with which the compound which has ether linkage and a bridge formation machine contains the compound over which the bridge was constructed in the bridge formation machine was used, while being able to obtain high membrane formation nature and strong film intensity again according to the rechargeable battery concerning the gestalt of this operation, high ion conductivity can also be obtained. Therefore, it can manufacture easily and a high performance can be obtained.

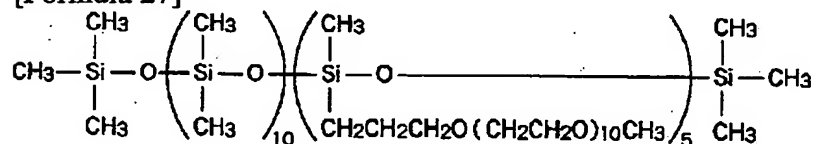
[0073]

[Example] Furthermore, the concrete example of this invention is explained in detail.

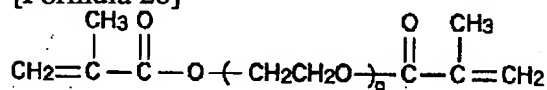
[0074] (Example 1) First, as a solvent, to the siloxane derivative and this siloxane derivative of 1 weight section shown in \*\* 27, 1.0 mols [ /kg ] lithium screw (trifluoromethyl sulfonyl) imide (Li (CF<sub>3</sub> SO<sub>2</sub>)<sub>2</sub> N) was added to the acetonitrile (CH<sub>3</sub> CN) of 50 weight sections, and it dissolved in it. Subsequently, the polyethylene glycol dimethacrylate of 2 weight sections shown in \*\* 28 as a compound which can construct a bridge over this, and the methoxy polyethylene glycol monomethacrylate of 2 weight sections shown in \*\* 29 were added, and it mixed until it became a uniform solution.

[0075]

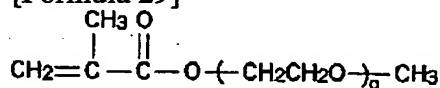
[Formula 27]



[Formula 28]



[Formula 29]



[0076] Then, after removing an acetonitrile until it kept at 25 degrees C under reduced pressure of this mixed solution and became 15 weight sections, add 2 of the 0.05 weight section, 2-dimethoxy -2, and a 2'-phenyl acetophenone as a polymerization initiator, and it was made to dissolve, and applied uniformly on the substrate made from Teflon. After applying on a substrate, it sets in 25-degree C air, and they are ultraviolet rays 6 mW/cm<sup>2</sup> It irradiated for 20 minutes with the quantity of light, and polymerization reaction was performed. It was made to dry after it and under reduced pressure of 60 degrees C for 6 hours, and the electrolyte with a thickness of 100 micrometers was obtained.

[0077] It is the film of this electrolyte an area of 0.7854cm<sup>2</sup> It clipped in the shape of a disk, it put between the electrode made from the stainless steel of a couple, and asked for the ionic conductivity in 25 degrees C by the alternating current impedance method. The result is shown in Table 1.

[0078]

[Table 1]

	架橋可能な化合物の添加量 (重量部)			イオン 伝導度 (mS/cm)
	化28に示した ジメタクリレー ト化合物	化29に示した モノメタクリレ ート化合物	化30に示した トリアクリレー ト化合物	
実施例1	2	2	—	0.15
2	0.5	0.5	—	0.22
3	0.125	0.125	—	0.43
4	1.33	2.66	—	0.23
5	0.83	0.166	—	0.51
6	—	—	4	0.016
7	—	—	1	0.087
8	—	—	0.25	0.31

[0079] (Example 2) Except for having made into the 0.5 weight section the addition of the polyethylene glycol dimethacrylate shown in-izing 28, and having made into the 0.5 weight section the addition of the methoxy polyethylene glycol monomethacrylate shown in \*\* 29, the electrolyte was produced like the example 1. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

[0080] (Example 3) Except for having made into the 0.125 weight section the addition of the polyethylene glycol dimethacrylate shown in-izing 28, and having made into the 0.125 weight section the addition of the methoxy polyethylene glycol monomethacrylate shown in \*\* 29, the electrolyte was produced like the example 1. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

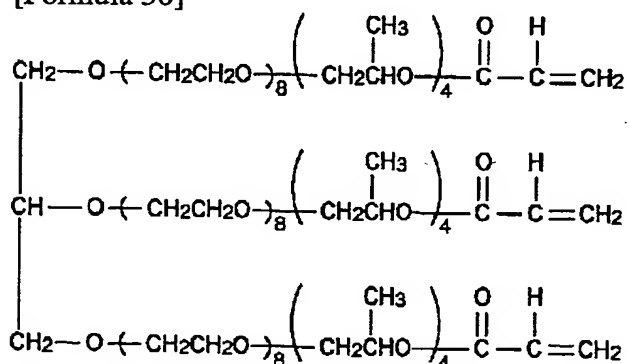
[0081] (Example 4) Except for having made into the 1.33 weight section the addition of the polyethylene glycol dimethacrylate shown in-izing 28, and having made into the 2.66 weight sections the addition of the methoxy polyethylene glycol monomethacrylate shown in \*\* 29, the electrolyte was produced like the example 1. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

[0082] (Example 5) Except for having made into the 0.083 weight section the addition of the polyethylene glycol dimethacrylate shown in-izing 28, and having made into the 0.166 weight section the addition of the methoxy polyethylene glycol monomethacrylate shown in \*\* 29, the electrolyte was produced like the example 1. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

[0083] (Example 6) the thoria chestnut rate compound which replaced with the methoxy polyethylene glycol monomethacrylate shown in the polyethylene glycol dimethacrylate shown in-izing 28, and \*\* 29, and was shown in \*\* 30 as a compound which can construct a bridge -- 4 weight \*\*\*\*\* -- except for things, the electrolyte was created like the example 1 Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

[0084]

[Formula 30]



[0085] (Example 7) the thoria chestnut rate compound which replaced with the methoxy polyethylene  
[http://www4.ipdl.jpo.go.jp/cgi-bin/tran\\_web.cgi\\_ejje](http://www4.ipdl.jpo.go.jp/cgi-bin/tran_web.cgi_ejje) 6/4/2003

glycol monomethacrylate shown in the polyethylene glycol dimethacrylate shown in-izing 28, and \*\* 29, and was shown in \*\* 30 -- 1 weight \*\*\*\*\* -- except for things, the electrolyte was created like the example 1 Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

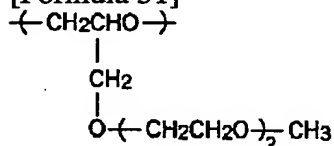
[0086] (Example 8) the thoria chestnut rate compound which replaced with the methoxy polyethylene glycol monomethacrylate shown in the polyethylene glycol dimethacrylate shown in-izing 28, and \*\* 29, and was shown in \*\* 30 -- 0.25 weight \*\*\*\*\* -- except for things, the electrolyte was created like the example 1 Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

[0087] (Example 9) the number of mols of the lithium screw imide to the number of mols of an ether linkage unit [ in / a siloxane derivative / for the siloxane derivative and lithium screw imide which were first shown in \*\* 27 ] -- it mixed so that it might be set comparatively (the number of mols of the ether linkage unit in the number of mols / siloxane derivative of lithium salt) to 0.06, and the mixture of a siloxane derivative was adjusted

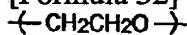
[0088] Subsequently, 20.6 mol % of composition units shown in \*\* 31, and 77.5 mol % of composition units shown in \*\* 32, The number average molecular weight which consists of 1.9 mol % of composition units shown in \*\* 33 prepares the solid-state-like random copolymer of 820,000 as a compound which can construct a bridge. It mixes so that it may be set comparatively (the number of mols of the ether linkage unit in the number of mols / copolymer of lithium salt) to 0.06. the number of mols of the lithium screw imide to the number of mols of an ether linkage unit [ in / a copolymer / for this copolymer and lithium screw imide ] -- The mixture of a copolymer was adjusted.

[0089]

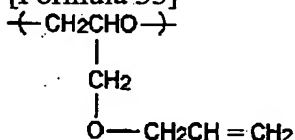
[Formula 31]



[Formula 32]



[Formula 33]



[0090] Then, the mixture of a siloxane derivative and the mixture of a copolymer were mixed in the acetonitrile so that the weight ratio (the mixture of a siloxane derivative mixture of a /copolymer) of the mixture of a siloxane derivative to the mixture of a copolymer might become 5/5, 2 and 2-dimethoxy -2 and the 2'-phenyl acetophenone were added as a polymerization initiator, and it considered as the mixed solution.

[0091] After adjusting a mixed solution, this mixed solution is uniformly applied on the substrate made from Teflon, and reduced pressure drying is carried out at 25 degrees C, and it sets in 25-degree C air, and they are ultraviolet raysmW [ 6 //cm ] 2 It irradiated for 20 minutes with the quantity of light, and polymerization reaction was performed. It was made to dry after it and under reduced pressure of 60 degrees C for 6 hours, and the electrolyte with a thickness of 100 micrometers was obtained. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is shown in Table 2.

[0092]

[Table 2]

	混合比 (重量比) シロキサン誘導体の混合物/共重合体の混合物	イオン 伝導度 (mS/cm)
実施例 9	5/5	0.156
10	2.5/7.5	0.385
11	7.5/2.5	0.0958
比較例	0/1	0.0103

[0093] (Example 10) Except for having mixed the mixture of a siloxane derivative, and the mixture of a copolymer so that the weight ratio (the mixture of a siloxane derivative mixture of a /copolymer) of the mixture of a siloxane derivative to the mixture of a copolymer might become 7.5/2.5, the electrolyte was produced like the example 9. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is shown in Table 2.

[0094] (Example 11) Except for having mixed the mixture of a siloxane derivative, and the mixture of a copolymer so that the weight ratio (the mixture of a siloxane derivative mixture of a /copolymer) of the mixture of a siloxane derivative to the mixture of a copolymer might become 2.5/7.5, the electrolyte was produced like the example 9. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is shown in Table 2.

[0095] (Example of comparison) Except for having not mixed the mixture of a siloxane derivative but having used only the mixture of a copolymer as an example of comparison over examples 9-11, the electrolyte was produced like the example 9. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is shown in Table 2.

[0096] It turns out that it has sufficient conductivity to obtain the ionic conductivity exceeding  $1 \times 10^{-5}$  S/cm from the result of examples 1-8 and examples 9-11, and use each electrolyte of an example for a cell from it. Moreover, the electrolyte of an example is also understood that high ionic conductivity is obtained compared with the example of comparison from the result of examples 9-11 and the example of comparison. That is, when the compound which has ether linkage and a bridge formation machine contained the compound over which the bridge was constructed in the bridge formation machine, and high molecular compounds, such as a siloxane derivative, ionic conductivity could be raised and this electrolyte was used, it turns out that the cell which has the outstanding performance is obtained.

[0097] As mentioned above, although the gestalt and each example of operation were given and this invention was explained, this invention is not limited to the gestalt and each example of the above-mentioned implementation, and can deform variously. For example, although the case where the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed was made to construct a bridge mutually was explained, you may make it make the compound which has ether linkage and for which a bridge can be constructed, and other compounds construct a bridge in the gestalt of the above-mentioned implementation. Namely, the compound with which the bridge was constructed over the compound which has ether linkage and a bridge formation machine in the bridge formation machine should just have at least the structure where the bridge was constructed over the compound which has ether linkage and a bridge formation machine, in part.

[0098] Moreover, as a compound which has the functional group in which ether linkage and bridge formation are possible in the gestalt of the above-mentioned implementation, although the copolymer including the composition unit shown in the ester compound shown in \*\* 19, \*\* 20, and \*\* 21 and the composition unit shown in \*\* 22, and \*\* 23 was explained concretely, if it has the functional group in which ether linkage and bridge formation are possible, what has other structures can be used similarly.

[0099] Furthermore, in the gestalt and each example of the above-mentioned implementation, although the siloxane derivative was explained as a high molecular compound, other high molecular compounds which can dissolve an electrolyte salt can be used similarly. In that case, you may make it use two or more high molecular compounds, and it may replace with a siloxane derivative, or other high molecular compounds may be used with a siloxane derivative.

[0100] In addition, in the gestalt of the above-mentioned implementation, although the rechargeable battery using the lithium was explained and the case where lithium salt was used as an electrolyte salt was explained in each above-mentioned example, this invention is applicable about the electrolyte and rechargeable battery which used other electrolyte salts, such as sodium salt or a calcium salt, similarly.

[0101] Furthermore, in the gestalt of the above-mentioned implementation, although the paper type rechargeable battery was explained, this invention is applicable about the thing of other configurations, such as telescopic [ which has a button type, a coin type, a square shape, or spiral structure ], similarly again.

[0102] In addition, although the case where the electrolyte of this invention was used for a rechargeable battery was explained in the gestalt of the above-mentioned implementation again, the constituent for an electrolyte and electrolytes of this invention can be used about other cells, such as a primary cell. Furthermore, it can also use for other electrochemistry devices, such as a capacitor, a capacitor, or an electrochromic element. In addition, for example, when using the electrolyte of this invention for a capacitor etc., basic-group salts, such as an ammonium salt, can also be used as an electrolyte salt.

[0103]

[Effect of the Invention] Since it was made for the compound and high molecular compound which have the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed to be included according to the constituent for electrolytes given in any 1 term of a claim 1 or a claim 16 as explained above, the electrolyte of this invention can be easily obtained by the polymerization. That is, the effect that the electrolyte which has high membrane formation nature and high ion conductivity can be obtained easily is done so.

[0104] Moreover, since it made the compound which has at least one free end which the functional group which can construct a bridge is not allotted as a compound which can construct a bridge, and includes ether linkage included according to the claim 3, the claim 7, the claim 8, or the constituent for electrolytes according to claim 10, the free end side chain which includes ether linkage by the polymerization can form, and the effect that the electrolyte which has higher ionic conductivity can obtain does so.

[0105] Furthermore, since it was made for the compound which has ether linkage and a bridge formation machine to contain the compound and high molecular compound over which the bridge was constructed in the bridge formation machine according to the electrolyte given in any 1 term of a claim 17 or a claim 32, a high molecular compound and an electrolyte salt can be held between the three-dimensions network structure. Therefore, ion conductivity can be made high, holding high membrane formation nature and strong film intensity. Therefore, if an electrochemistry device is constituted using this electrolyte, the effect that the electrochemistry device which has a high performance easily can be obtained will be done so.

[0106] In addition, since it was made to have the free end side chain in which the compound over which the bridge was constructed includes ether linkage according to the electrolyte according to claim 19 Moreover, since it was made to have the structure where the bridge was constructed over the monoester compound expressed with \*\* 8 according to the electrolyte according to claim 23 or 24 Moreover, since it was made to have the structure where the bridge was constructed over the copolymer including the composition unit shown in \*\* 10 in the bridge formation machine according to the electrolyte according to claim 26, the effect that higher ionic conductivity can be obtained is done so.

[0107] Furthermore, since it was made to carry out the polymerization of the compound for which a bridge can be constructed again according to the manufacture method of an electrolyte given in any 1 of a claim 33 or the claims 36 after mixing the compound and high molecular compound which have the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, the effect that the electrolyte of this invention can be obtained easily is done so.

[0108] In addition, since the electrolyte of this invention was used, while being able to obtain high membrane formation nature and strong film intensity again according to the cell given in any 1 term of a claim 37 or a claim 52, high ion conductivity can also be obtained. Therefore, it can manufacture



easily and the effect that a high performance can be obtained is done so.

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[Translation done.]



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TECHNICAL FIELD

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[The technical field to which invention belongs] this invention relates to the cell which used an electrolyte, its manufacture method, and it for the constituent row for electrolytes containing a high molecular compound and an electrolyte salt.

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**PRIOR ART**

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[Description of the Prior Art] Recent years come, portable electrical-and-electric-equipment products, such as a camcorder/movie (video tape recorder), a cellular phone, or a laptop computer, are spreading quickly, and the further high performance-ization of an electrochemistry device is needed as a device of such electronic equipment.

[0003] Conventionally, the liquefied electrolyte made to dissolve an electrolyte salt in water or an inflammable organic solvent has been used as matter with which electrochemistry devices, such as a rechargeable battery, manage ionic conduction. However, a liquefied electrolyte has problems, such as a liquid spill, and, for this reason, needed to secure confidentiality using the metal container.

Therefore, generally, such an electrochemistry device had the heavy weight, and the flexibility of a configuration also had complicatedness low [ a device ] in connection with the sealing process. Then, the research using the so-called solid electrolyte which consists of an ion conductivity solid-state is made actively. While there are no worries about a liquid spill in a solid electrolyte and being able to carry out simple [ of the sealing process for it ], there is an advantage, like the flexibility of shape selectivity is high by the film moldability which can carry out [ lightweight ]-izing of the device and which was further excellent in the polymer.

[0004] Generally this solid electrolyte consists of a matrix macromolecule and a possible electrolyte salt of ionic dissociation. Among these, the matrix macromolecule holds ionic dissociation ability and has the function of both sides, such as a duty which solidifies this ion conductivity solid-state, and a duty as a solvent to an electrolyte salt. About such a solid electrolyte, the report that the ionic conductivity of about  $1 \times 10^{-7}$  S/cm was obtained by the system which dissolved the lithium perchlorate in the polyethylene oxide by Armand and others of the Grenoble university (France) is carried out in 1978, and examination has also been performed still more actively since then about the various polymeric materials focusing on the polymer which has polyether combination now.

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## EFFECT OF THE INVENTION

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[Effect of the Invention] Since it was made for the compound and high molecular compound which have the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed to be included according to the constituent for electrolytes given in any 1 term of a claim 1 or a claim 16 as explained above, the electrolyte of this invention can be easily obtained by the polymerization. That is, the effect that the electrolyte which has high membrane formation nature and high ion conductivity can be obtained easily is done so.

[0104] Moreover, since it was made the compound which has at least one free end which the functional group which can construct a bridge is not allotted as a compound which can construct a bridge, and includes ether linkage to be included according to the claim 3, the claim 7, the claim 8, or the constituent for electrolytes according to claim 10, the free end side chain which includes ether linkage by the polymerization can form, and the effect that the electrolyte which has higher ionic conductivity can be obtained does so.

[0105] Furthermore, since it was made for the compound which has ether linkage and a bridge formation machine to contain the compound and high molecular compound over which the bridge was constructed in the bridge formation machine according to the electrolyte given in any 1 term of a claim 17 or a claim 32, a high molecular compound and an electrolyte salt can be held between the three-dimensions network structure. Therefore, ion conductivity can be made high, holding high membrane formation nature and strong film intensity. Therefore, if an electrochemistry device is constituted using this electrolyte, the effect that the electrochemistry device which has a high performance easily can be obtained will be done so.

[0106] In addition, it is since it was made to have the free end side chain in which the compound over which the bridge was constructed includes ether linkage according to the electrolyte according to claim 19. Moreover, since it was made to have the structure where the bridge was constructed over the monoester compound expressed with \*\* 8 according to the electrolyte according to claim 23 or 24, and since it was made to have the structure where the bridge was constructed over the copolymer including the composition unit shown in \*\* 10 in the bridge formation machine according to the electrolyte according to claim 26, the effect that higher ionic conductivity can be obtained is done so.

[0107] Furthermore, since it was made to carry out the polymerization of the compound for which a bridge can be constructed again according to the manufacture method of an electrolyte given in any 1 of a claim 33 or the claims 36 after mixing the compound and high molecular compound which have the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, the effect that the electrolyte of this invention can be obtained easily is done so.

[0108] In addition, since the electrolyte of this invention was used, while being able to obtain high membrane formation nature and strong film intensity again according to the cell given in any 1 term of a claim 37 or a claim 52, high ion conductivity can also be obtained. Therefore, it can manufacture easily and the effect that a high performance can be obtained is done so.

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## TECHNICAL PROBLEM

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[Problem(s) to be Solved by the Invention] The solid electrolyte using a straight chain-like polyether which is represented by this polyethylene oxide as a matrix shows ionic conduction, when the ion which is dissolving moves by the local segmental motion of a macromolecule chain in the inside of the amorphous phase more than the glass transition temperature of a matrix polymer.

[0006] However, the ion which is dissolving into the matrix of the shape of a straight chain like the polyethylene oxide which is the polymer of semicrystallinity, especially the cation were strongly configured according to the interaction with a macromolecule chain, this became a point constructing [ false ] a bridge, and they caused partial crystallization, and had the problem that a segmental motion will fall. Therefore, in order to realize higher ionic conductivity under a room temperature, to perform a molecular design which the dissociating power of an electrolyte salt is high, and the amorphous (amorphous) field where ion tends to move within a matrix exists mostly, and keeps the glass transition point of a polymer low is desired.

[0007] In addition, the attempt which raises ionic conductivity by introducing branching structure into a polyethylene-oxide frame as this kind of a molecular design, for example is performed (21 the Watanabe justice, Netsu Sokutei 24 (1) pp12- 1996). However, this kind of polymer has the problem that the synthetic method is complicated.

[0008] Moreover, otherwise, the three-dimensions network structure is introduced into a matrix macromolecule, the attempt which makes crystallization of a polymer check is made, and the method of carrying out the polymerization of the acrylic or the methacrylic system monomer which has a polyoxyalkylene component as an example of this kind of molecular design is reported (JP,5-25353,A). However, since the melting nature to the monomer of an alkali-metal salt is low, there is a problem that sufficient ionic conductivity is not obtained. Therefore, the new solid electrolyte replaced with these is called for.

[0009] this invention was made in view of this trouble, and the purpose is in offering the cell which used an electrolyte, its manufacture method, and it for the constituent row for electrolytes which can show high ion conductivity.

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MEANS

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[Means for Solving the Problem] The constituent for electrolytes by this invention contains the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt.

[0011] The compound with which the electrolyte by this invention has ether linkage and a bridge formation machine contains the compound over which the bridge was constructed in the bridge formation machine, a high molecular compound, and an electrolyte salt.

[0012] The manufacture method of the electrolyte by this invention mixes the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt, and makes the compound which can construct a bridge construct a bridge.

[0013] After the manufacture method of other electrolytes by this invention mixes the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, and a high molecular compound and makes the compound which can construct a bridge construct a bridge, it adds an electrolyte salt.

[0014] The cell by this invention is equipped with an electrolyte with a positive electrode and a negative electrode, and an electrolyte contains the compound with which the bridge was constructed over the compound which has ether linkage and a bridge formation machine in the bridge formation machine, a high molecular compound, and an electrolyte salt.

[0015] In the constituent for electrolytes by this invention, since the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, and the high molecular compound are included, the electrolyte in which high ion conductivity is shown is obtained.

[0016] In the electrolyte by this invention, the electrolyte salt dissociated with the high molecular compound between the compounds with which the bridge was constructed over the compound which has ether linkage and a bridge formation machine in the bridge formation machine exists. Therefore, high ion conductivity is shown.

[0017] By the manufacture method of the electrolyte by this invention, after the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt are mixed, a bridge is constructed over the compound for which a bridge can be constructed.

[0018] By the manufacture method of the electrolyte by this invention, after the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, and a high molecular compound are mixed and the polymerization of the compound which can construct a bridge is carried out, an electrolyte salt is added.

[0019] By the cell by this invention, when the ion produced by the maceration of an electrolyte salt moves between a negative electrode and a positive electrode in the inside of an electrolyte, it discharges. Here, since it has the electrolyte of this invention, it has high ion conductivity and the outstanding cell performance is shown.

[0020]

[Embodiments of the Invention] Hereafter, the gestalt of operation of this invention is explained in detail.

[0021] The electrolyte concerning the gestalt of 1 operation of this invention contains the compound

which has the three-dimensions network structure by which the bridge was constructed over the compound which has ether linkage and a bridge formation machine in the bridge formation machine, the high molecular compound, and the electrolyte salt. Thus, the compound which has the three-dimensions network structure and over which the bridge was constructed is included because membranes cannot be formed since a fluidity is shown as molecular weight becomes low, and it is hard to obtain sufficient film intensity only with a high molecular compound also in a high molecular weight object. That is, the compound which has this three-dimensions network structure and over which the bridge was constructed is for holding a high molecular compound and an electrolyte salt between the three-dimensions network structure, and collateralizing membrane formation nature and strong film intensity.

[0022] As this compound over which the bridge was constructed, what has the structure where the bridge was constructed over either [ at least ] the straight chain-like compound or the compound of the pectinate in the bridge formation machine is mentioned, and you may have the side chain, for example. In addition, this compound over which the bridge was constructed is not restricted to that by which the bridge was constructed over the open chain compound that what is necessary is just to have the structure where the bridge was constructed over the compound which contains a chain-like atomic arrangement in part at least for example, but may have the structure where the bridge was constructed over the compound which has an annular atomic arrangement in part. That is, this compound over which the bridge was constructed may have the annular atomic arrangement in the principal chain or the side chain.

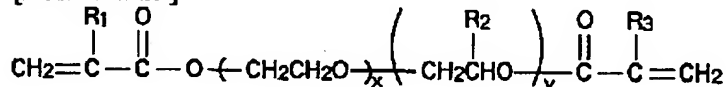
[0023] Moreover, a bridge may be constructed over the compound with which the compound with which this compound over which the bridge was constructed has at least one bridge formation machine has a bridge formation machine in what position that what is necessary is just to construct a bridge. For example, although the bridge was constructed over the straight chain-like compound, the bridge may be constructed over the compound which has a bridge formation machine to ends, and may be constructed over the compound which has a bridge formation machine at one end at the case. Although the bridge was constructed over the compound of the pectinate, the bridge may be constructed over the compound which has a bridge formation machine at at least one end at the case. Namely, this compound over which the bridge was constructed should just form the three-dimensions network structure which can hold a high molecular compound and an electrolyte salt.

[0024] However, in order to realize higher ionic conductivity, it is desirable to have the structure where the bridge was constructed over the compound which has at least one free end which a bridge formation machine is not arranged and includes ether linkage. It is because it will have the free end side chain which branching structure is introduced into the three-dimensions network structure by this, and includes ether linkage. For example, when constituted by the compound with which the bridge was constructed over the straight chain-like compound, it is desirable to have the structure where the bridge was constructed over the compound which has a bridge formation machine at one end in addition to the structure where the bridge was constructed over the compound which has a bridge formation machine to ends. Moreover, when constituted by the compound with which the bridge was constructed over the compound of the pectinate, it is desirable to have the structure where the bridge was constructed over the compound of the pectinate which has at least one free end on which a bridge formation machine is not arranged. Furthermore, when constituted by the compound with which the bridge was constructed over the straight chain-like compound and the compound of the pectinate, it is desirable to have the structure where the bridge was constructed over the compound of the shape of a straight chain which has a bridge formation machine at one end, or the structure where the bridge was constructed over the compound of the pectinate which has at least one free end which is not matched with a bridge formation machine.

[0025] This compound over which the bridge was constructed may have the structure of cross linkage by combination like a throat, such as ether linkage, ester combination, or a urethane bond, again. For example, the compound which has an allyl group, a vinyl group, an acrylate machine, a methacrylate machine, the functional group that has cycloolefin structure, or an active hydrogen machine may have the structure of cross linkage over which the bridge was constructed in them. Moreover, you may have the structure of cross linkage from which plurality differs in 1 compound.

[0026] Specifically as such a compound over which the bridge was constructed, what has the structure where what has the structure of cross linkage by which the bridge was constructed over the ester compound in the bridge formation machine was mentioned, and the bridge was constructed over at least one sort in a monoester compound, a diester compound, and a triester compound especially is desirable. For example, what has the structure where the bridge was constructed over the diester compound including the oxy-alkylene structure expressed with the general formula shown in \*\* 19 as a compound which has the structure where the bridge was constructed over the diester compound in the bridge formation machine is mentioned.

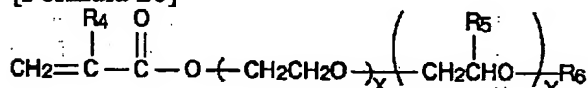
[Formula 19]



The inside of a formula, R1, and R2 And R3 A hydrogen atom or the alkyl group of carbon numbers 1-6 is expressed, and even if each is the same, it may differ. Moreover, x and y are the integers of  $x \geq 1$  and  $y \geq 0$  or  $x \geq 0$ , and  $y \geq 1$ .

[0027] In this case, it is more desirable to have the structure where the bridge was constructed over the monoester compound with which the diester compound expressed with the general formula shown in \*\* 19, for example includes the oxy-alkylene structure which is expressed with the general formula shown in \*\* 20 in addition to the structure where the bridge was constructed in the bridge formation machine in the bridge formation machine. It is because a free end side chain including ether linkage is introduced and higher ionic conductivity can be obtained, as mentioned above.

[Formula 20]

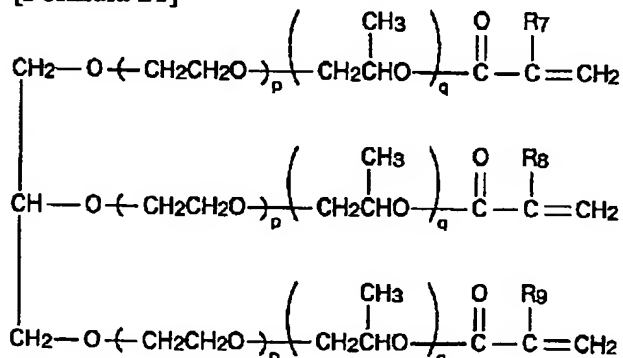


The inside of a formula, R4, and R5 And R6 A hydrogen atom or the alkyl group of carbon numbers 1-6 is expressed, and even if each is the same, it may differ. Moreover, X and Y are the integers of  $X \geq 1$  and  $Y \geq 0$  or  $X \geq 0$ , and  $Y \geq 1$ .

[0028] In this case, as for the structure where the bridge was constructed over the diester compound in the bridge formation machine, and the structure where the bridge was constructed over the monoester compound in the bridge formation machine, in the weight ratio (a monoester compound / diester compound) of a monoester compound to a diester compound, it is desirable that it is within the limits of 5.0 or less more greatly than 0. It is because the three-dimensions network structure cannot be constituted if there is little structure where the bridge was constructed over the diester compound, but the free end side chain which includes ether linkage when there is little structure where the bridge was constructed over the monoester compound decreases and ionic conductivity cannot be made high.

[0029] Moreover, what has the structure where the bridge was constructed over the triester compound including the oxy-alkylene structure expressed with the general formula shown in \*\* 21 as a compound which has the structure where the bridge was constructed over the triester compound, for example in the bridge formation machine is mentioned.

[Formula 21]



The inside of a formula, R7, and R8 And R9 A hydrogen atom or the alkyl group of carbon numbers 1-6 is expressed, and even if each is the same, it may differ. Moreover, p and q are the integers of  $p \geq 1$  and  $q \geq 0$  or  $p \geq 0$ , and  $q \geq 1$ .

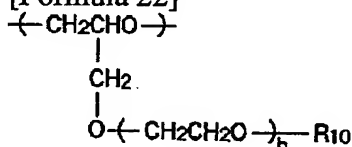
[0030] Although especially the composition ratio of the oxyethylene unit of the triester compound shown in this \*\* 21 and an oxypropylene unit is not limited, it is desirable that it is within the limits of  $0.1 \leq q/p \leq 4$ . It is because it will become weak if many [ if there are too few oxyethylene units, a mechanical strength will fall, and / too ]. Moreover, although any of the shape of the letter of a block or random are sufficient as the joint format of an oxyethylene unit and an oxypropylene unit, the shape of random is more desirable.

[0031] It is more desirable to have the structure where the bridge was constructed over the monoester compound expressed with the general formula shown in \*\* 20 in the bridge formation machine in addition to the structure where the bridge was constructed over the triester compound expressed with the general formula shown in \*\* 21 in the bridge formation machine, as mentioned above also in this case.

[0032] moreover -- as the compound over which the bridge was constructed -- these -- others -- for example, what has the structure where the bridge was constructed over the copolymer including the composition unit shown in the composition unit shown in \*\* 22 and \*\* 23 in the bridge formation machine is mentioned Since according to this compound the free end side chain which includes ether linkage by the composition unit shown in \*\* 22 is introduced and high ionic conductivity can be obtained, it is desirable.

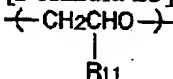
[0033]

[Formula 22]



R10 expresses at least one sort in the alkyl group of carbon numbers 1-12, the alkenyl machine of carbon numbers 2-18, the cycloalkyl machine of carbon numbers 3-8, the aryl group of carbon numbers 6-18, the aralkyl machine of carbon numbers 7-12, and a tetrahydropyranyl group among a formula, and h is the integer of  $1 \leq h \leq 12$ . In addition, R10 may express the same content in all composition units, and may express the content which changes with composition units.

[Formula 23]



R11 expresses among a formula at least one sort in at least one side of the functional groups which have at least one side of the functional groups which have an active hydrogen machine and an unsaturation double bond or an active hydrogen machine, and an unsaturation double bond, a hydrogen atom, an alkyl group, an alkenyl machine, a cycloalkyl machine, and an aryl group. In addition, R11 may express the same content in all composition units, and may express the content which changes with composition units.

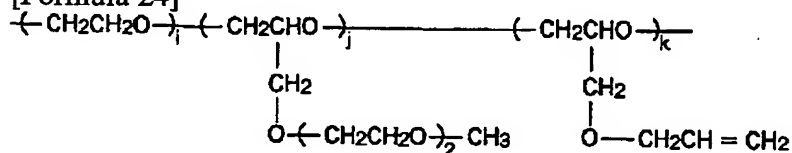
[0034] Any of the shape of the letter of a block or random are sufficient as the joint format with the composition unit shown in the composition unit which especially the composition ratio with the composition unit shown in the composition unit shown in \*\* 22 in this copolymer and \*\* 23 is not limited, and was shown in \*\* 22, and \*\* 23. Incidentally, in this copolymer, the functional group which can construct a bridge is a functional group which has an active hydrogen machine and an unsaturation double bond. As a functional group which has an unsaturation double bond, the functional group which has an allyl group, a vinyl group, an acrylate machine, a methacrylate machine, or cycloolefin structure is mentioned.

[0035] That by which the bridge was constructed over the copolymer specifically expressed with the general formula shown in \*\* 24 as a compound which has such structure, and over which the bridge was constructed in the bridge formation machine is desirable. In \*\* 22, as for the copolymer shown in



\*\* 24, k includes [R10 / R11 ] the composition unit which consists of an allyl group in the composition unit which consists of a methyl group, the composition unit which R11 becomes from a hydrogen atom in \*\* 23, and \*\* 23 by two.

[Formula 24]



i, j, and k are one or more integers among a formula.

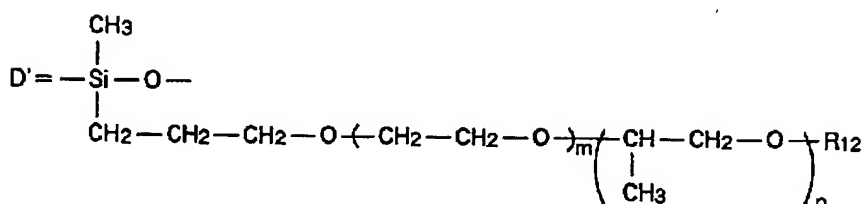
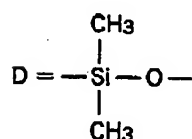
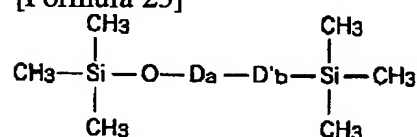
[0036] In addition, you may mix and use two or more compounds with which such structures differ for this compound over which the bridge was constructed. For example, you may mix and use the compound which has the structure where the bridge was constructed over the copolymer including the compound which has the structure where the bridge was constructed over the ester compound shown in \*\* 19, and the composition unit shown in the composition unit shown in \*\* 22, and \*\* 23.

[0037] A high molecular compound is for dissolving an electrolyte salt. It is n (SiOCH<sub>3</sub> R) which has the chain combination of silicon (Si) and oxygen (O) in a basic skeleton and by which the side chain R was added to silicon as this high molecular compound, for example. The shape type siloxane derivative of a chain expressed is mentioned. In this case, a substituent or a side chain radical R is a monovalent organic machine, and serves as the structure where an electrolyte salt can be dissolved, by choosing a side chain radical R moderately. Therefore, as for the average molecular weight of this siloxane derivative, it is desirable that it is 100000 or less.

[0038] Moreover, since high compatibility with the compound with which the bridge was constructed over the compound which has ether linkage in the bridge formation machine is needed while dissolving an electrolyte salt effectively, as for this siloxane derivative, it is desirable to include ether linkage in a substituent or a side chain radical R. As such a side chain radical R, there are alkoxy groups, such as an ethoxy basis, a propoxy group, a butoxy machine, and a methoxy machine. Moreover, the hydrogen in a side chain radical R may be replaced by halogens, such as boron or a fluorine.

[0039] What is specifically expressed with the general formula shown in \*\* 25 as such a siloxane derivative is desirable.

[Formula 25]



a expresses the integer of 0 to 100 among a formula, b expresses the integer of 1 to 100, m expresses the integer of 0 to 100, n expresses the integer of 0 to 100, and R<sub>12</sub> expresses a hydrogen atom or an alkyl group. In addition, at the time of b > 1, even if b D' is the same, they may differ. Moreover, the hydrogen atom contained in D' and R<sub>12</sub> may be replaced by the halogen atom.

[0040] An electrolyte salt is for giving ion conductivity by maceration, and is expressed with the general formula shown in \*\* 26.

[Formula 26] A expresses a cation among AB formula and B expresses an anion.

[0041] A light metal salt etc. is mentioned as this electrolyte salt, and, specifically, it is used according to the purpose from alkaline-earth-metals salts, such as alkali-metal salts, such as a lithium (Li) salt, a sodium (Na) salt, or a potassium (K) salt, a calcium (calcium) salt, or a magnesium (Mg) salt, etc. In addition As lithium salt \*\*  $\text{LiClO}_4$ ,  $\text{LiBF}_4$ ,  $\text{LiPF}_6$ ,  $\text{LiAsF}_6$ ,  $\text{LiAlCl}_4$ ,  $\text{LiSbF}_6$ ,  $\text{LiCF}_3\text{SO}_3$ ,  $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ ,  $\text{LiC}_4\text{F}_9\text{SO}_3$ ,  $\text{LiCF}_3\text{CO}_2$ , and  $\text{LiN}(\text{CF}_3\text{CO}_2)_2$  It is. etc. -- as sodium salt --  $\text{NaClO}_4$ ,  $\text{NaBF}_4$ ,  $\text{NaSCN}$ , etc. -- it is -- as potassium salt --  $\text{KBF}_4$  etc. -- it is The need is accepted, a gap, one sort, or two sorts or more are mixed, and these are used.

[0042] In addition, when a siloxane derivative is used as a high molecular compound, as for the rate of the compound and high molecular compound over which the bridge was constructed, it is desirable to make into within the limits below the 10000 weight sections the compound over which the bridge was constructed to the 100 weight sections of a siloxane derivative more than 10 weight sections. It is because un-arranging -- a film becomes easily or hard -- will arise if film intensity is not enough if there are few compounds over which the bridge was constructed than 10 weight sections, and the 10000 weight sections are exceeded, although based also on the molecular weight of a siloxane derivative, or an electrolytic use.

[0043] Moreover, the rate of the electrolyte salt to the compound over which the bridge was constructed is a rate (the number of mols of the ether linkage unit in the number of mols / compound with which the bridge was constructed of an electrolyte salt) of the number of mols of an electrolyte salt to the number of mols of the ether linkage unit in the compound over which the bridge was constructed, and it is desirable that it is within the limits of five or less [ 0.0001 or more ]. As for the rate of the electrolyte salt to a siloxane derivative, it is desirable similarly that it is [ of the number of mols of the electrolyte salt to the number of mols of the ether linkage unit in a siloxane derivative ] within the limits of five or less [ 0.0001 or more ] comparatively (the number of mols of the ether linkage unit in the number of mols / siloxane derivative of an electrolyte salt). It is because high ionic conductivity can be obtained within the limits of this.

[0044] The number of mols of the ether linkage unit incidentally included in one mol of diester compounds shown in \*\* 19 is mol  $(1+x+y)$ , and the number of mols of the ether linkage unit included in one mol of copolymers shown in \*\* 24 is  $\{i+(1+1+2)xj+(1+1)xk\}$  mol.

[0045] The electrolyte which has such composition can be manufactured as follows using the following constituents for electrolytes.

[0046] First, the constituent for electrolytes containing the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt is prepared. Among these, a high molecular compound and an electrolyte salt are as having mentioned above. For example, a siloxane derivative is prepared as a high molecular compound, and at least one sort of lithium salt is prepared as an electrolyte salt.

[0047] The compound which can construct a bridge turns into a compound which has the three-dimensions network structure mentioned above according to bridge formation and over which the bridge was constructed, the compounds or those mixture of the pectinate are sufficient also as a straight chain-like compound, and they may have the side chain. Moreover, as long as it has the chain-like atomic arrangement in part at least, the compound which has an atomic arrangement also with an open chain compound annular to a part of principal chain or side chain is sufficient. Furthermore, if it has at least one functional group which can construct a bridge, any number of numbers are good and the position is also good anywhere. For example, in the case of the straight chain-like compound, you may have to ends the functional group which can construct a bridge, and may have only at one end. You may be the many organic functions which may have the functional group which can construct a bridge over at least one end in the case of the compound of the pectinate, and it has three or more.

[0048] Moreover, the compound which can construct a bridge may have what functional group as a functional group which can construct a bridge. Specifically, the compound which has an ester

compound or an allyl group is mentioned. For example, as long as it is an ester compound, you may have a monoester compound, a diester compound, a triester compound, or ester combination four or more. Furthermore, the compound in which this bridge formation is possible may contain two or more sorts of things from which the number of the functional groups which can construct a bridge differs, and may contain two or more sorts of things from which the kind of functional group which can construct a bridge differs.

[0049] The triester compound expressed with the general formula shown in the monoester compound and \*\* 21 which are expressed with the diester compound expressed with the general formula shown in \*\* 19 as a compound in which such bridge formation is possible, for example, and the general formula shown in \*\* 20 is mentioned. In addition, it is more desirable to include the monoester compound shown in \*\* 20 in addition to this compound, when using the diester compound shown in \*\* 19. As mentioned above, when this constructs a bridge, it is for forming a free end side chain including ether linkage. In this case, as for the weight ratio (a monoester compound / diester compound) of a monoester compound to a diester compound, it is desirable that it is within the limits of 5.0 or less more greatly than 0. Moreover, it is more desirable to include the monoester compound shown in \*\* 20 in addition to this compound, when using the triester compound shown in \*\* 21.

[0050] A copolymer including the composition unit shown in the composition unit shown in \*\* 22 and \*\* 23 as a compound which can construct a bridge over others, for example etc. is mentioned. Specifically, the copolymer shown in \*\* 24 is desirable.

[0051] In addition, it is desirable to make into within the limits below the 10000 weight sections the compound which can construct a bridge more than 10 weight sections to the 100 weight sections of a siloxane derivative as the rate of the compound and high molecular compound which can construct a bridge was mentioned above, when a siloxane derivative was used as a high molecular compound. Moreover, the rate of the electrolyte salt to the compound which can construct a bridge as mentioned above The number of mols of the electrolyte salt to the number of mols of the ether linkage unit in the compound which can construct a bridge comparatively (the number of mols of the ether linkage unit in the compound in which electrolytic number of mols / bridge formation are possible) The rate of the aforementioned electrolyte salt [ as opposed to / it is desirable that it is within the limits of five or less / 0.0001 or more / , and / a siloxane derivative ] It is desirable that it is [ of the number of mols of the electrolyte salt to the number of mols of the ether linkage unit in a siloxane derivative ] within the limits of five or less [ 0.0001 or more ] comparatively (the number of mols of the ether linkage unit in electrolytic number of mols / siloxane derivative).

[0052] Subsequently, the compound which has the functional group in which these ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt are mixed, and uniform mixture is produced. In addition, a polymerization initiator is also added if needed in that case. A polymerization initiator is needed except when irradiation of an electron ray performs a polymerization, and it has a photopolymerization initiator and a thermal polymerization initiator.

[0053] As a photopolymerization initiator, there is acetophenone, TORIKURORO acetophenone, 2-hydroxy-2-methylpropiohenone, 2-hydroxy-2-methyl iso propiohenone, 1-hydroxy cyclohexyl ketone, BENZO iso ether, 2, and 2-diethoxy acetophenone or a benzyl dimethyl ketal, for example. Moreover, as a thermal polymerization initiator, there is initiators, such as elevated-temperature polymerization initiators, such as cumene hydroperoxide, t-butyl hydroperoxide, a JIKUMI loopier oxide, or G t-butyl peroxide, or a benzoyl peroxide, a lauroyl peroxide, a persulfate, or an azobisisobutyronitril, or a redox initiator, for example. As a polymerization initiator, you may mix and use 1 of sorts of these, and two sorts or more.

[0054] In addition, a polymerization initiator is added in the range of the 0.1 - 1.0 weight section to the compound 100 weight section which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed. It is because an effect does not change even if a rate of polymerization will fall remarkably and will add exceeding the 1.0 weight sections, if fewer than the 0.1 weight section.

[0055] Then, ultraviolet rays, an electron ray, X-rays, gamma rays, microwave, or a RF is irradiated at this mixture, or this mixture is heated, and the polymerization of the compound which can construct a

bridge is carried out. A bridge is constructed over the compound which can construct a bridge by this, and the electrolyte concerning the gestalt of this operation is obtained.

[0056] In addition, in case the compound which can construct a bridge, a high molecular compound, and an electrolyte salt are mixed, proper organic solvents, such as an acetonitrile, are used and you may make it form mixture. In this case, after performing dryness processing of leaving or heating in air and under reduced pressure after carrying out the polymerization of the compound which can construct a bridge, and making it remove an organic solvent, and obtaining the mixture, performing dryness processing and removing an organic solvent, you may be made to carry out the polymerization of the compound which can construct a bridge.

[0057] Moreover, this electrolyte can be manufactured even if it performs it as follows.

[0058] First, the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed, a high molecular compound, and an electrolyte salt are prepared like the previous manufacture method. Subsequently, it mixes with the compound which can construct a bridge, and a high molecular compound. A polymerization initiator is mixed if needed in that case. Then, the polymerization of the compound which can construct a bridge is carried out like the previous manufacture method, and the compound which can construct a bridge forms the mixture of the compound and high molecular compound over which the bridge was constructed in the bridge formation machine. Dissolve an electrolyte salt in the organic solvent after it, this mixture is made to permeate, and dryness removal of the organic solvent is carried out.

Thereby, the electrolyte concerning the gestalt of this operation is obtained.

[0059] In addition, although especially the conditions of polymerization reaction are not limited, polymerization reaction is considered from the simple nature of equipment, and a cost side, and UV irradiation or its heating polymerization is desirable. Moreover, when, producing the electrolyte used for the lithium cell using alkali-metal ion, a lithium ion battery, and a sodium cell for example, as for the solvent and polymerization initiator at the time of mixing the constituent for electrolytes (namely, the compound, high molecular compound, and alkali-metal salt which can construct a bridge), and these, it is desirable to fully perform dehydration processing, and it is desirable to also make atmosphere under manufacture into low humidity. It is because there are some which are decomposed depending on the kind of alkali-metal salt when this reacts with moisture. Moreover, it is because alkali metal and moisture react violently in a negative electrode when it uses as a cell.

[0060] Thus, the electrolyte manufactured is used for a cell as follows. Here, the example of the rechargeable battery which used the lithium is given and it explains below with reference to a drawing.

[0061] Drawing 1 expresses the cross-section structure of the rechargeable battery using the electrolyte concerning the gestalt of this operation. In addition, what was shown in drawing 1 is called so-called paper type. The laminating of this rechargeable battery is carried out through the electrolyte 13 which requires a negative electrode 11 and a positive electrode 12 for the gestalt of this operation. sheathing of the couple which the insulating packing 14 is arranged by these sides, and was arranged so that it might face across a negative electrode 11 and a positive electrode 12 in between -- it is sealed by pasting up with the periphery section of members 15 and 16

[0062] The negative electrode 11 contains occlusion and the metal which can be \*\*\*\*ed, the alloy, or the carbon material for for example, the lithium metal or the lithium ion. This carbon material is prepared in predetermined temperature and predetermined atmosphere, for example, carbon black, such as corks, such as pyrolytic carbons, petroleum coke, or pitch coke, artificial graphites, natural graphites, and acetylene black, glassy carbons, the organic polymeric-materials baking object, or the carbon fiber is used. In addition, an organic polymeric-materials baking object calcinates organic polymeric materials at the suitable temperature of 500 degrees C or more in inert gas atmosphere or a vacuum.

[0063] a positive electrode 12 -- as a positive active material --  $\text{TiS}_2$ ,  $\text{MoS}_2$ , and  $\text{NbSe}_2$  Or  $\text{V}_2\text{O}_5$  etc. -- the metallic sulfide which does not contain a lithium, the oxide, the lithium compound sulfide containing a lithium, or the lithium multiple oxide is contained In order to make an energy density high especially, it is  $\text{Li}_x\text{MO}_2$ . It is desirable that the lithium multiple oxide made into a subject is included. In addition, one or more kinds of transition metals of M are desirable, and, specifically, its

at least one sort in cobalt (Co), nickel (nickel), and manganese (Mn) is desirable. Moreover,  $x$  is usually the value of  $0.05 \leq x \leq 1.10$  within the limits. as the example of such a lithium multiple oxide --  $\text{LiCoO}_2$ ,  $\text{LiNiO}_2$ ,  $\text{Li}_x\text{Ni}_y\text{Co}_{1-y}\text{O}_2$ , or (however, the value of  $x$  and  $y$  changes with charge-and-discharge states of a cell, and are usually  $0 < x < 1$  and  $0.7 < y \leq 1$ .)  $\text{LiMn}_2\text{O}_4$  etc. -- it is mentioned [0064] In addition, this lithium multiple oxide carries out trituration mixture according to composition of a request of the carbonate of a lithium, a nitrate, an oxide or a hydroxide, and the carbonate of transition metals, a nitrate, an oxide or a hydroxide, and is prepared by calcinating in oxygen atmosphere at the temperature within the limits of 600-1000 degrees C.

[0065] The electrolyte 13 contains lithium salt as an electrolyte salt here. Moreover, the electrolyte 13 serves also as the duty as separator in this rechargeable battery. That is, a negative electrode 11 and a positive electrode 12 are isolated, and a lithium ion is passed, preventing the short circuit of the current by contact of two poles. In addition, you may make it have the separator which is not illustrated between a positive electrode 12 and an electrolyte 13 or into an electrolyte 13 between a negative electrode 11 and an electrolyte 13 if needed. As separator, there is what consists of a nonwoven fabric made of synthetic resin, such as a polytetrafluoroethylene, polypropylene, or polyethylene, a thing which consists of a ceramic film, or a thing which consists of a porosity thin film film.

[0066] The rechargeable battery which has such composition acts as follows.

[0067] In this rechargeable battery, if it charges, a lithium will serve as ion, it will be desorbed from a positive electrode 12, and occlusion will be carried out to a negative electrode 11 through an electrolyte 13, for example. If it discharges, a lithium serves as ion and it is desorbed from a negative electrode 11, and through an electrolyte 13, a metaphor will return to a positive electrode 12 and occlusion will be carried out. Here, the compound which has ether linkage and a bridge formation machine contains the compound over which the bridge was constructed in the bridge formation machine, and the electrolyte 13 holds the high molecular compound and the lithium ion between the three-dimensions network structure. Therefore, while having membrane formation nature and strong film intensity, it also has high ion conductivity. Therefore, it has the outstanding cell performance.

[0068] Thus, since it was made for the compound which has ether linkage and a bridge formation machine to contain the compound and high molecular compound over which the bridge was constructed in the bridge formation machine according to the electrolyte concerning the gestalt of this operation, a high molecular compound and an electrolyte salt can be held between the three-dimensions network structure. Therefore, ion conductivity can be made high, holding high membrane formation nature and strong film intensity. Therefore, if an electrochemistry device is constituted using this electrolyte, the electrochemistry device which has a high performance easily can be obtained.

[0069] Moreover, if it is made to have the free end side chain in which the compound over which the bridge was constructed includes ether linkage, higher ionic conductivity can be obtained.

[0070] Furthermore, since it was made for the compound and high molecular compound which have the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed to be included according to the constituent for electrolytes concerning the gestalt of this operation, the polymerization of the compound in which this bridge formation is possible can be easily carried out by irradiation or heating of ultraviolet rays. Therefore, the electrolyte easily built over the gestalt of this operation can be obtained.

[0071] In addition, since it was made to carry out the polymerization of the compound for which a bridge can be constructed after mixing the compound and high molecular compound which have the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed according to the manufacture method of the electrolyte concerning the gestalt of this operation, the electrolyte easily built over the gestalt of this operation can be obtained.

[0072] Furthermore, since the electrolyte with which the compound which has ether linkage and a bridge formation machine contains the compound over which the bridge was constructed in the bridge formation machine was used, while being able to obtain high membrane formation nature and strong film intensity again according to the rechargeable battery concerning the gestalt of this operation, high ion conductivity can also be obtained. Therefore, it can manufacture easily and a high

performance can be obtained.

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[Translation done.]

## \* NOTICES \*

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2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

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**EXAMPLE**

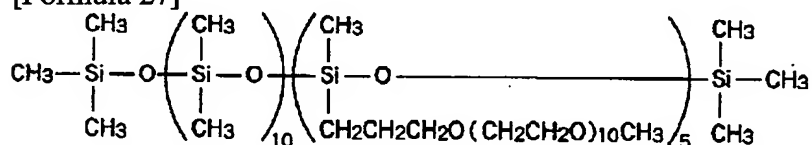

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[Example] Furthermore, the concrete example of this invention is explained in detail.

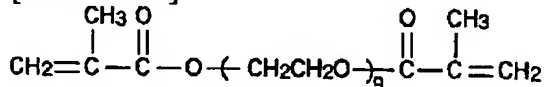
[0074] (Example 1) First, as a solvent, to the siloxane derivative and this siloxane derivative of 1 weight section shown in \*\* 27, 1.0 mols [ /kg ] lithium screw (trifluoromethyl sulfonyl) imide (Li (CF<sub>3</sub> SO<sub>2</sub>)<sub>2</sub> N) was added to the acetonitrile (CH<sub>3</sub> CN) of 50 weight sections, and it dissolved in it. Subsequently, the polyethylene glycol dimethacrylate of 2 weight sections shown in \*\* 28 as a compound which can construct a bridge over this, and the methoxy polyethylene glycol monomethacrylate of 2 weight sections shown in \*\* 29 were added, and it mixed until it became a uniform solution.

[0075]

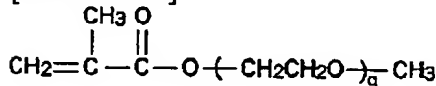
[Formula 27]



[Formula 28]



[Formula 29]



[0076] Then, after removing an acetonitrile until it kept at 25 degrees C under reduced pressure of this mixed solution and became 15 weight sections, add 2 of the 0.05 weight section, 2-dimethoxy -2, and a 2'-phenyl acetophenone as a polymerization initiator, and it was made to dissolve, and applied uniformly on the substrate made from Teflon. After applying on a substrate, it sets in 25-degree C air, and they are ultraviolet rays 6 mW/cm<sup>2</sup> It irradiated for 20 minutes with the quantity of light, and polymerization reaction was performed. It was made to dry after it and under reduced pressure of 60 degrees C for 6 hours, and the electrolyte with a thickness of 100 micrometers was obtained.

[0077] It is the film of this electrolyte an area of 0.7854cm<sup>2</sup> It clipped in the shape of a disk, it put between the electrode made from the stainless steel of a couple, and asked for the ionic conductivity in 25 degrees C by the alternating current impedance method. The result is shown in Table 1.

[0078]

[Table 1]

	架橋可能な化合物の添加量 (重量部)			イオン 伝導度 (mS/cm)
	化28に示した ジメタクリレー ト化合物	化29に示した モノメタクリレ ート化合物	化30に示した トリアクリレー ト化合物	
実施例1	2	2	—	0.15
2	0.5	0.5	—	0.22
3	0.125	0.125	—	0.43
4	1.33	2.66	—	0.23
5	0.83	0.166	—	0.51
6	—	—	4	0.016
7	—	—	1	0.067
8	—	—	0.25	0.31

[0079] (Example 2) Except for having made into the 0.5 weight section the addition of the polyethylene glycol dimethacrylate shown in-izing 28, and having made into the 0.5 weight section the addition of the methoxy polyethylene glycol monomethacrylate shown in \*\* 29, the electrolyte was produced like the example 1. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

[0080] (Example 3) Except for having made into the 0.125 weight section the addition of the polyethylene glycol dimethacrylate shown in-izing 28, and having made into the 0.125 weight section the addition of the methoxy polyethylene glycol monomethacrylate shown in \*\* 29, the electrolyte was produced like the example 1. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

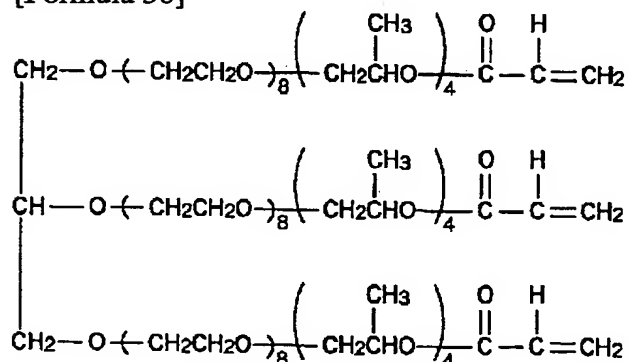
[0081] (Example 4) Except for having made into the 1.33 weight section the addition of the polyethylene glycol dimethacrylate shown in-izing 28, and having made into the 2.66 weight sections the addition of the methoxy polyethylene glycol monomethacrylate shown in \*\* 29, the electrolyte was produced like the example 1. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

[0082] (Example 5) Except for having made into the 0.083 weight section the addition of the polyethylene glycol dimethacrylate shown in-izing 28, and having made into the 0.166 weight section the addition of the methoxy polyethylene glycol monomethacrylate shown in \*\* 29, the electrolyte was produced like the example 1. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

[0083] (Example 6) the thoria chestnut rate compound which replaced with the methoxy polyethylene glycol monomethacrylate shown in the polyethylene glycol dimethacrylate shown in-izing 28, and \*\* 29, and was shown in \*\* 30 as a compound which can construct a bridge -- 4 weight \*\*\*\*\* -- except for things, the electrolyte was created like the example 1 Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

[0084]

[Formula 30]



[0085] (Example 7) the thoria chestnut rate compound which replaced with the methoxy polyethylene  
[http://www4.ipdl.jpo.go.jp/cgi-bin/tran\\_web.cgi\\_ejje](http://www4.ipdl.jpo.go.jp/cgi-bin/tran_web.cgi_ejje) 6/4/2003



glycol monomethacrylate shown in the polyethylene glycol dimethacrylate shown in-izing 28, and \*\* 29, and was shown in \*\* 30 -- 1 weight \*\*\*\*\* -- except for things, the electrolyte was created like the example 1 Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

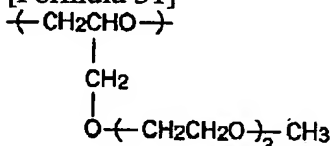
[0086] (Example 8) the thoria chestnut rate compound which replaced with the methoxy polyethylene glycol monomethacrylate shown in the polyethylene glycol dimethacrylate shown in-izing 28, and \*\* 29, and was shown in \*\* 30 -- 0.25 weight \*\*\*\*\* -- except for things, the electrolyte was created like the example 1 Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is similarly shown in Table 1.

[0087] (Example 9) the number of mols of the lithium screw imide to the number of mols of an ether linkage unit [ in / a siloxane derivative / for the siloxane derivative and lithium screw imide which were first shown in \*\* 27 ] -- it mixed so that it might be set comparatively (the number of mols of the ether linkage unit in the number of mols / siloxane derivative of lithium salt) to 0.06, and the mixture of a siloxane derivative was adjusted

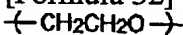
[0088] Subsequently, 20.6 mol % of composition units shown in \*\* 31, and 77.5 mol % of composition units shown in \*\* 32, The number average molecular weight which consists of 1.9 mol % of composition units shown in \*\* 33 prepared the solid-state-like random copolymer of 820,000 as a compound which can construct a bridge, and it mixed so that it might be set comparatively (the number of mols of the ether linkage unit in the number of mols / copolymer of lithium salt) to 0.06, and the mixture of the mol number of lithium screw imide to the number of mols of an ether linkage unit [ in / a copolymer / for this copolymer and lithium screw imide ] which is an

[0089]

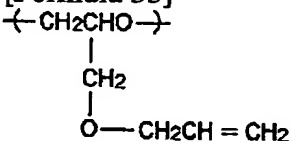
[Formula 31]



[Formula 32]



[Formula 33]



[0090] Then, the mixture of a siloxane derivative and the mixture of a copolymer were mixed in the acetonitrile so that the weight ratio (the mixture of a siloxane derivative mixture of a /copolymer) of the mixture of a siloxane derivative to the mixture of a copolymer might become 5/5, 2 and 2-dimethoxy -2 and the 2'-phenyl acetophenone were added as a polymerization initiator, and it considered as the mixed solution.

[0091] After adjusting a mixed solution, this mixed solution is uniformly applied on the substrate made from Teflon, and reduced pressure drying is carried out at 25 degrees C, and it sets in 25-degree C air, and they are ultraviolet raysmW [ 6 //cm ] 2 It irradiated for 20 minutes with the quantity of light, and polymerization reaction was performed. It was made to dry after it and under reduced pressure of 60 degrees C for 6 hours, and the electrolyte with a thickness of 100 micrometers was obtained. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is shown in Table 2.

[0092]

[Table 2]

	混合比 (重量比) シロキサン誘導体の混合体／共重合体の混合体	イオン 伝導度 (mS/cm)
実施例 9	5/5	0.158
10	2.5/7.5	0.385
11	7.5/2.5	0.0958
比較例	0/1	0.0103

[0093] (Example 10) Except for having mixed the mixture of a siloxane derivative, and the mixture of a copolymer so that the weight ratio (the mixture of a siloxane derivative mixture of a /copolymer) of the mixture of a siloxane derivative to the mixture of a copolymer might become 7.5/2.5, the electrolyte was produced like the example 9. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is shown in Table 2.

[0094] (Example 11) Except for having mixed the mixture of a siloxane derivative, and the mixture of a copolymer so that the weight ratio (the mixture of a siloxane derivative mixture of a /copolymer) of the mixture of a siloxane derivative to the mixture of a copolymer might become 2.5/7.5, the electrolyte was produced like the example 9. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is shown in Table 2.

[0095] (Example of comparison) Except for having not mixed the mixture of a siloxane derivative but having used only the mixture of a copolymer as an example of comparison over examples 9-11, the electrolyte was produced like the example 9. Ionic conductivity was investigated about this electrolyte as well as an example 1. The result is shown in Table 2.

[0096] It turns out that it has sufficient conductivity to obtain the ionic conductivity exceeding  $1 \times 10^{-5}$  S/cm from the result of examples 1-8 and examples 9-11, and use each electrolyte of an example for a cell from it. Moreover, the electrolyte of an example is also understood that high ionic conductivity is obtained compared with the example of comparison from the result of examples 9-11 and the example of comparison. That is, when the compound which has ether linkage and a bridge formation machine contained the compound over which the bridge was constructed in the bridge formation machine, and high molecular compounds, such as a siloxane derivative, ionic conductivity could be raised and this electrolyte was used, it turns out that the cell which has the outstanding performance is obtained.

[0097] As mentioned above, although the gestalt and each example of operation were given and this invention was explained, this invention is not limited to the gestalt and each example of the above-mentioned implementation, and can deform variously. For example, although the case where the compound which has the functional group in which ether linkage and bridge formation are possible and for which a bridge can be constructed was made to construct a bridge mutually was explained, you may make it make the compound which has ether linkage and for which a bridge can be constructed, and other compounds construct a bridge in the gestalt of the above-mentioned implementation. Namely, the compound with which the bridge was constructed over the compound which has ether linkage and a bridge formation machine in the bridge formation machine should just have at least the structure where the bridge was constructed over the compound which has ether linkage and a bridge formation machine, in part.

[0098] Moreover, as a compound which has the functional group in which ether linkage and bridge formation are possible in the gestalt of the above-mentioned implementation, although the copolymer including the composition unit shown in the ester compound shown in \*\* 19, \*\* 20, and \*\* 21 and the composition unit shown in \*\* 22, and \*\* 23 was explained concretely, if it has the functional group in which ether linkage and bridge formation are possible, what has other structures can be used similarly.

[0099] Furthermore, in the gestalt and each example of the above-mentioned implementation, although the siloxane derivative was explained as a high molecular compound, other high molecular compounds which can dissolve an electrolyte salt can be used similarly. In that case, you may make it use two or more high molecular compounds, and it may replace with a siloxane derivative, or other high molecular compounds may be used with a siloxane derivative.

[0100] In addition, in the gestalt of the above-mentioned implementation, although the rechargeable battery using the lithium was explained and the case where lithium salt was used as an electrolyte salt was explained in each above-mentioned example, this invention is applicable about the electrolyte and rechargeable battery which used other electrolyte salts, such as sodium salt or a calcium salt, similarly.

[0101] Furthermore, in the gestalt of the above-mentioned implementation, although the paper type rechargeable battery was explained, this invention is applicable about the thing of other configurations, such as telescopic [ which has a button type, a coin type, a square shape, or spiral structure ], similarly again.

[0102] In addition, although the case where the electrolyte of this invention was used for a rechargeable battery was explained in the gestalt of the above-mentioned implementation again, the constituent for an electrolyte and electrolytes of this invention can be used about other cells, such as a primary cell. Furthermore, it can also use for other electrochemistry devices, such as a capacitor, a capacitor, or an electrochromic element. In addition, for example, when using the electrolyte of this invention for a capacitor etc., basic-group salts, such as an ammonium salt, can also be used as an electrolyte salt.

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[Translation done.]

\* NOTICES \*

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## DESCRIPTION OF DRAWINGS

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[Brief Description of the Drawings]

[Drawing 1] It is a cross section showing the composition of the rechargeable battery using the electrolyte concerning the gestalt of 1 operation of this invention.

[Description of Notations]

11 -- negative electrode, 12 -- positive electrode, and 13 -- an electrolyte, 14 -- insulation packing, 15, and 16 -- sheathing -- a member

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[Translation done.]

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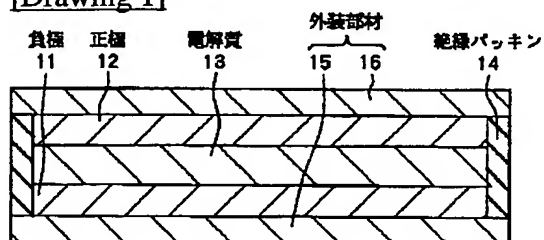
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DRAWINGS

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[Drawing 1]



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